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EVALUATION OF ADIABATIC REFORMER IN MIXED-GAS-CYCLE

FINAL TECHNICAL REPORT (UNCLASSIFIED)

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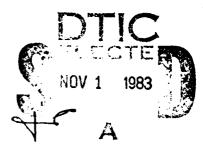
Contract DAAK-70-82-C-0012

25 June 1983

Power Systems Division
UNITED TECHNOLOGIES CORPORATION

Prepared for

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EVALUATION OF ADIABATIC REFORMER IN MIXED-GAS-CYCLE

FINAL REPORT

Contract DAAK70-82-C-0012

J. A. S. Bett R. R. Lesieur A. P. Meyer E. C. Swift L. L. Van Dine

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SUMMARY

The work under this program investigated two different approaches to fuels processing for military fuel cell power plants. One process was evaluated relative to the requirements for a tactical family of power plants, specifically, the ability to operate on high sulfur logistic fuels content, potential for multi-fuel operation, and freedom from the need for water supply or water recovery. The second process, also for operation on sulfur-containing logistic fuel, had as its key evaluation criteria high efficiency and reliability. This process was to be used in power plants located at fixed sites where fuel and maintenance support transportation costs represent a significant portion of the power plant's life-cycle-cost. Other factors that weighed heavily in the selection were development status and development cost.

The process for tactical power plant applications, adiabatic reforming in a mixed-gas-cycle, was shown to be a basis for meeting the requirements of this application. Thermodynamic analyses established an operating regime and performance goals for the reformer as used in this process. Tests of a two-pound per-hour reactor (2-kW equivalent) showed that operation in this regime was satisfactory. Operation on sulfur-containing diesel fuel, methanol, and contaminated methanol were demonstrated. Work in a previous program demonstrated unleaded gasoline would also be a suitable fuel.

The test program included analysis of the product gas composition, determination of the effect of impurities in the fuel cell exhaust on reactor performance, and the effect of total pressure on reactor performance. Tests at low pressure were conducted with a fuel injection nozzle especially developed for this application.

Based on the test data, the weight, volume, and parasite power requirements for the members of a power plant family from 1.5 to 100-kW were determined. The process was found to be preferred for power plants of 10-kW rating or higher. Above 10-kW the effect on weight to provide the benefit of multi-logistic fuel capability was not great.

The process selected as the preferred choice for remote site applications was cyclic reforming. The process was selected by comparison of four alternative processes. The four processes were Thermal Steam Reforming, Adiabatic Reforming, Hybrid Reforming, and Cyclic Reforming. Each of these techniques had been previously studied by United both analytically and experimentally for commercial service. This background served as a base for these studies.

Intrinsic capability comparisons were made of the concepts. One was rejected. Systems based on each of the remaining fuel processing methods were established. Comparisons of estimated efficiency, cost and reliability were made.

Their current development status and the development each required for reduction to practice were also defined and evaluated. With this data the preferred process was selected.

Cyclic reforming was judged to best meet the overall evaluation criteria for ease of supply and service at a remote site and development status and cost. Further evaluation of cyclic reforming was recommended and a development plan for carrying forward evaluation of this process was defined.

SECTION 1

INTRODUCTION

Fuel cell power plants for military applications must often meet different operating requirements than those developed for commercial service. An example of this is the capability to operate on logistic fuels with relatively high sulfur levels. Another, for reasons of mobility, is low weight. Environmental requirements may impose freezing problems on a power plant, making liquid water in the power plant unacceptable. Still other applications for remote permanent sites, where logistic support is costly or difficult, require high efficiency to minimize fuel supply and reliability to minimize maintenance support.

How well a fuel cell power plant meets these requirements is significantly affected by the choice of fuel conditioning system incorporated into the power plant. Under this program the choice of fuel conditioning system was examined for two different military applications. The first was for a family of tactical power plants and the second was for a remote site application.

TACTICAL APPLICATION POWER PLANTS

The key requirements for the fuels processor of these power plants were (1) to be able to operate on logistically available fuels and (2) to be free from the problems associated with freeze protection. A fuel processor based on the adiabatic reformer UTC developed for the Electric Power Research Institute was proposed for this application. It had demonstrated the ability to operate on high sulfur No. 2 oil in EPRI sponsored tests (1) and on high sulfur diesel oil and unleaded gasoline under U.S. Army sponsored work, Contract DAAK70-80-C-0115. However both of these efforts had a system concept which required a liquid water supply.

Under this program the adiabatic reformer was incorporated into a system referred to as a mixed-gas-cycle in which no liquid water supply is required. The water required in the reforming process is instead supplied as a vapor directly from the cathode exhaust.

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These system changes met the power plant operating requirements but changed the operating conditions previous programs had verified for the adiabatic reformer. In the mixed-gas-cycle system the adiabatic reformer runs water-lean. The effect of impurities in the cathode exhaust stream would have to be evaluated as would the potential for ammonia formation. Ammonia may form in this system because nitrogen is brought into the reformer via the cathode exhaust. If significant ammonia forms, it must be scrubbed from the fuel gas since ammonia adversely reacts with phosphoric acid electrolytes.

Similarly, changes occurred in physical operating conditions. Direct use of cathode exhaust requires the reformer to be run close to ambient pressure which required evaluation of a low-pressure reformer fuel nozzle.

Systems analyses were conducted to determine a probable regime for reformer operation in the mixed-gas-cycle.

Tests were conducted of the reformer in this regime and the effects of acid and nitrogen in the reformer feed were determined. The reformer was tested as well with a low pressure nozzle.

Finally, weight, volume, and efficiency estimates were made for a family of power plants, based on the mixed-gas-cycle system and the test experience, in the range of 1.5 to 100-kW.

REMOTE SITE POWER PLANT

The remote site application studied focused on the adaptation of a specific United fuel cell power plant, the 40-kW PC18, to a specific service. This application set a premium on overall electrical efficiency since fuel transportation cost comprised a significant portion of total life-cycle-cost. Minimized maintenance requirements similarly were desired because of the difficulty of site access.

Four alternative fuel processing methods were evaluated for adapting the commercial power plants for operation on logistic fuels at high efficiency. They were Thermal Steam Reforming, Adiabatic Reforming, Hybrid Reforming, and Cyclic Reforming. Each of these techniques had been previously studied by United both analytically and experimentally for commercial service. This background served as a base for these studies.

Intrinsic capability comparisons were made of the concepts. One was rejected. Systems based on each of the remaining fuel processing methods were established. Comparisons of estimated efficiency, cost and reliability were made.

A preferred system was selected and a development plan for that concept was defined.

REPORT FORMAT

This Final Report is divided into two major sections. The first deals with the work to investigate the Tactical Family of Power Plants. The second reviews the work done to define the preferred fuel processor for the remote site applications. Each section is a free standing entity and may be used without the need to refer to the other.

SECTION 2

INVESTIGATION OF ADIABATIC REFORMER IN MIXED-GAS-CYCLE FOR TACTICAL POWER PLANT APPLICATIONS

PROGRAM APPROACH

This work was carried out in several subtasks. These were: (1) Thermodynamic Analyses, (2) Reactor and Fuels Testing, (3) Data Analysis and (4) Power Plant Characteristics, Estimates and Analysis. The thermodynamic analyses were conducted to thermodynamically match an adiabatic reformer and a phosphoric acid stack into a mixed-gas-cycle power plant system and to determine the range of operating conditions representative of power plant operation under which to test the reactor.

The adiabatic reformer used for testing was a 2-inch diameter reactor containing United's advanced metal oxide and metal catalysts. It was calibrated with diesel fuel at conventional adiabatic reformer conditions of previous tests. The test stand was then modified to allow the reformer to operate at mixed-gas-cycle conditions. Both diesel fuel and methanol fuels were tested over a range of conditions likely to be required for Army mixed-gas-cycle operation. After the main body of data had been collected, a direct fuel injection nozzle was designed, built and tested in the adiabatic test reactor, and finally the effect of phosphoric acid addition which would be present from cathode exhaust was evaluated.

This body of data established the basis for a preliminary component definition to size and evaluate power plant performance for power plants with ratings of from 1.5 to 100-kW.

FUELS SELECTION

Three fuels were selected in conjunction with the Army for testing in this program. They were: diesel fuel, neat methanol and neat methanol contaminated with 5% diesel fuel. The diesel fuel was considered to be typical of Army logistic diesel

fuel, methanol was selected as a potential fuel for this type of power plant, and the contaminated methanol was equivalent to methanol which might be transferred to a power plant in a container which previously contained diesel fuel. A temperature of about 100°F was required to completely dissolve 5% diesel in methanol. A comparison of the properties of the diesel fuel used in the previous study (2) with the diesel fuel used in the present study, listed in Table 2-1 shows the two fuels to be very similar. Methanol was purchased to the AA grade methanol specification requiring a purity of 99.85 percent by methanol by weight. Table 2-2 is the AA grade methanol specification taken from Federal Specification O-H-232F.

TABLE 2-1. DIESEL FUEL PROPERTIES

	Previous Study	Present <u>Fuel</u>
Specific Gravity	0.853	0.858
H/C	1.73	1.71
Aromatics %v	36.0	33.0
Sulfur PPMw	1090	1300
Distillation °F		
I.B. Pt.	155	245
10%	408	450
50%	540	545
70%	587	580
90%	655	630
95%	680	650

TABLE 2-2. GRADE AA METHANOL SPECIFICATIONS

	Grade A		Grade A	4
Characteristics	Requirement	Test Para.	Requirement	Test Para.
Acetone and alde- hydes, percent max.	0.003	4.4.1.2	0.003	4.4.1.2
	!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!!	•	0 002	7 7 7
Ethanol, percent max.		1 1 1 1 1 1 1	20.0	4.4.
Acidity (as Acetic Acid) percent max	0.003	4.4.1.4	0.003	4.4.1.4
Appearance and hydro- carbons	Free of opalescence, suspended matter	4.4.1.5	Free of opalescence, suspended matter	4.4.1.5
Carbonizable sub-	and sediment Not darker than	4 4 1 6	and sediment	•
stances, Color	Color Standard No. 30 of ASTM D 1209, Platinum-		Not darker than Color Standard No. 30 of ASTM D 1209, Platinum-	4.4.1.6
Color	Cobail Scale		Cobalt Scale	
	Not darker than Color Standard No. 5 of ASTM D 1209, Platinum- Cobalt Scale	4.4.1.7	Not darker than Color Standard No. 5 of ASTM D 1209, Platinum- Cobalt Scale	4.4.1.7
Distillation range	Not more than 1°C and shall include 64.6°C ± .10°C at 760 mm	4.4,1.8	Not more than 1°C and shall include 64.6°C ± .10°C	4.4.1.8
Specific gravity, max. Percent methanol by	0.7928 at 20°/20°C 99.85	4.4.1.9	at /60 mm. 0.7928 at 20°/20°C	4.4.1.9
weight, min.			69.66	4.4.1.9
Nonvolatile content, gm/100 ml, max.	0.0010	4.4.1.10	0.0010	4.4.1.10
Odor	Characteristic, non-residual	4.4.1.11	Characteristic,	4.4.1.11
Permanganate	No discharge of Color in 30 minutes	4.4.1.12	No discharge of	4.4.1.12
Water, percent max.	0.15	4.4.1.13	Color in 30 minutes 0.10	4 4 1 13

THERMODYNAMIC ANALYSES TO ESTABLISH OPERATIONAL AND PERFORMANCE GOALS

This subtask defined the thermodynamic regime within which an adiabatic reformer operates in a mixed-gas-cycle on logistic fuels. It establishes a match between the interfaces of the fuel processor and the other subsystems of the power plant. With a match defined, the operating conditions of the reformer may be determined.

Description of Mixed-Gas-Cycle Concept

In the mixed-gas-cycle, moisture laden gas from the cathode vent of the fuel cell stack is mixed with the fuel stream that is injected into the adiabatic reformer. A summary of operating experience, characteristics and key operating requirements of adiabatic reformers is presented in Appendix A. The cathode vent gas provides the oxygen required to effect combustion in the adiabatic reformer and the water required for the reforming process. Because the cathode vent gas is added directly into the reformer, no external water supply is required nor need the water be recovered as a liquid and revised as is done in conventional fuel cell power plant reformer systems. The steam-to-carbon ratio in the feed to the reformer is dependent on the hydrogen utilization of the anode, and the oxygen used in the reformer is dependent on the oxygen utilization of the cathode.

The arrangement of an adiabatic reformer and phosphoric acid fuel cell stack in a mixed-gas-cycle is shown in Figure 2-1. Air is drawn into the system and is blown through the stack as well as through the burner used to preheat the reformer feed. As the air passes through the stack, it supplies the oxygen required for the electrochemical reaction and removes product water from the cells. The moisture laden oxygen-depleted air passes through heat exchanger No. I into the adiabatic reformer. Heat exchanger No. 1 preheats the reformer feed from the cathode exhaust to the temperature required to operate the adiabatic reformer with minimal oxygen addition by using the heat generated by burning excess hydrogen from the cell anode. In the reformer, the fuel is vaporized and combusted to raise the gas mixture to reforming temperatures. The heated gases then pass through

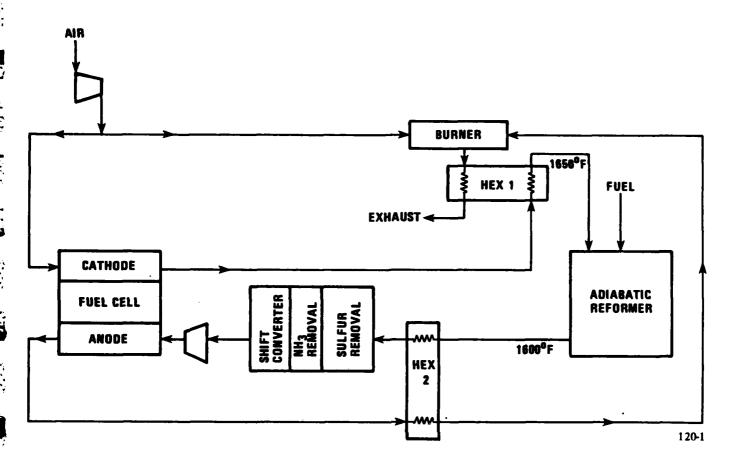


Figure 2-1. Schematic of Conceptual Power Plant

the reformer and become converted to a hydrogen rich stream. During this conversion, the gases cool as the reforming process proceeds.

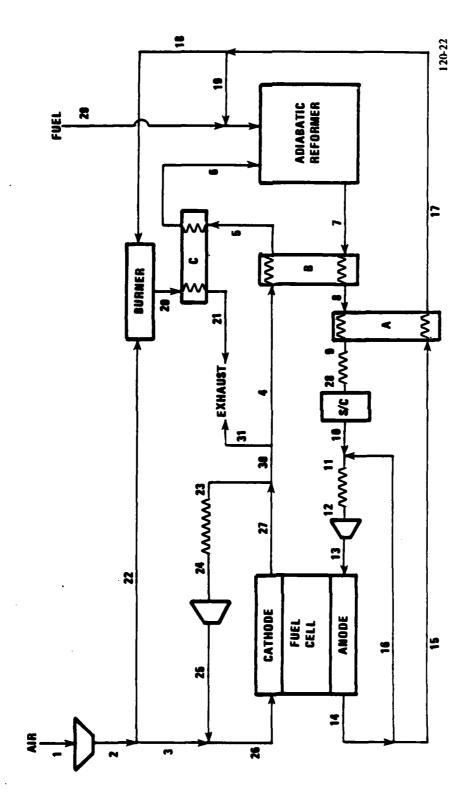
The hydrogen rich gas leaves the reformer and is cooled in heat exchanger No. 2 before being purified and upgraded in hydrogen content by a sulfur removal bed, an ammonia removal bed, and a shift converter which converts excess carbon monoxide formed in the reformer to hydrogen and carbon dioxide. The cooled, cleaned, upgraded gas passes through the anode to provide hydrogen for the electrochemical reaction. The unutilized hydrogen and inerts in the anode stream are then utilized as the coolant stream in heat exchanger No. 2, and then combusted to heat the adiabatic reformer feed stream.

General System Definition for Thermodynamic Analyses

A mixed-gas-cycle fuel cell power plant was defined which included all heat exchangers, blowers, pumps and gas purifiers, and which matched fuel and air requirements for both the reformer and the fuel cell. Several cell cooling options and operating parameters were included in the system to permit analytical investigation of options to optimize cell performance. Steam to carbon ratio and cell oxygen and hydrogen utilization were varied in order to optimize power plant efficiency.

The generalized mixed-gas-cycle power plant system used for the thermodynamic study is shown in Figure 2-2. It includes both anode and cathode coolant loops, a bleed at station 31 which would allow the cathode to run at a lower oxygen utilization while still maintaining the oxygen requirements of the reformer, anode recycle to the reformer, three regenerative type heat exchangers and several air cooled heat exchangers. Station numbers are assigned to identify the upstream and downstream side of each unit. A computer program was used to close material and energy balances around each unit and around the system.

A typical computer output showing temperatures, gas flows and enthalpies at each station is shown in Figure 2-3. This case is for a 1.5-kW power plant operating on diesel fuel. A diesel fuel flow rate, shown as No. 2, of 0.1044 pound moles per hour (1.44 pounds per hour) was required to produce 1.5-kW of net power. The fuel cell stack consisted of 80 cells operating at 340°F and 0.499 volts per cell. The reformer exit temperature, TATR, was set at 1600°F. Power plant operating time was assumed to be 6000 hours with 1336 starts. The HEX analysis gives important heat exchanger information for those regenerative heat exchangers being used in the system. In this typical case, HEX B was not used since it was a hot fuel to hot air heat exchanger, and its use was not considered to be good engineering practice. A series of these computer runs was made for both diesel and methanol fuels in order to obtain test conditions for the reformer which satisfied all components of the system.



Schematic of Generalized Mixed-Gas-Cycle Power Plant Figure 2-2.

	EFF= ****** HEX AN EFF=	84 344 6994 165 1696 29	1 9000 9090 1 4996 1 1506 1 15	8 1600 7 1450 JUA COL ELTH, DE IN- 0 2209 6 1652 JUA COL	<<< D- LTC- 2 >>> -	0 104 0 268 0 260 0 200 0 200 1 500 0 1 500 0 1 683 0 9 000 0 0 600 0 0 000 0 0 0 000 0 0 0 000 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0	200 000 000 000 000 000 000 000 000 000	.>>>> <<<< 1081.000 .>>>> <<<< 1232.606
12345678901234567890123456789017	77 7 9 9 9 9 9 9 9 9 9 9 9 9 9 9 9 9 9	0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0	NTU	00000000000000000000000000000000000000	COOOOOOOOOOOOOOOOOOOOOOOOOOOOOOOOOOOOO	1.748 C02 0.0 0.0 0.0 0.0 0.0 0.0 0.0 0.0 0.0 0	-5 083 02 5822 0 15829 0 11886 0 0386 0 0386 0 0 0	N2 EHTH 2809 30076 22294 -12274 0 440 -12274 0 440 -12274 0 488 -7664 4 0 488 -7664 0 488 -4437731 12129 -454627 12129 -454627 12129 -454627 12129 -454627 12129 -454627 12129 -454627 12129 -488 -11714 0 488 -11571 0 488 -11571 0 488 -11571 0 488 -11571 0 488 -11571 0 488 -11571 0 488 -11571 0 488 -11571 0 488 -11571 0 595 -16327 4 0 000 -11 0 000 0 000 0 000 0 000 0 000 0 000 0 0

Figure 2-3. Typical Computer Output from Mixed-Gas-Cycle System Study

A series of cases were run on the computer to define the steam to carbon ratio at which the reformer would have to be run while operating on diesel fuel. The oxygen to carbon ratio into the reformer was fixed at 0.37 moles of oxygen per mole of carbon, and the steam to carbon ratio (cell hydrogen utilization) was varied for cell voltages of 0.5, 0.55 and 0.6 volts. The results plotted in Figure 2-4 show that lowering the voltage decreases the number of cells per stack, it also shows that at each voltage level, there is a steam to carbon ratio which minimizes the number of cells per stack. An operating level of 0.5 volts per cell was selected for power plant systems evaluation. This resulted in a stack containing a

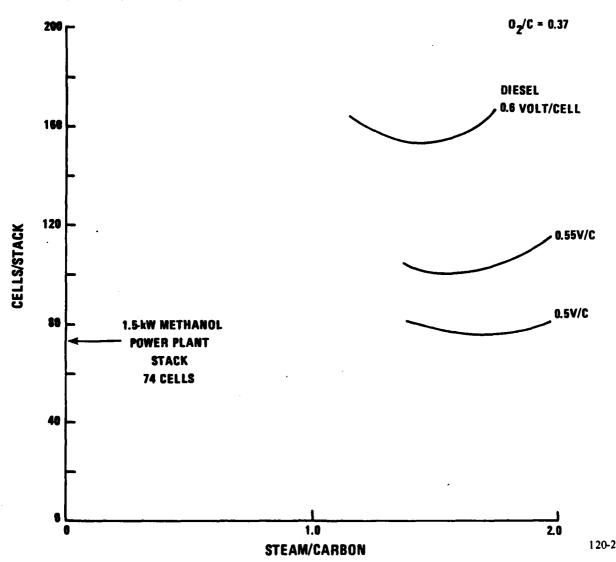


Figure 2-4. Steam to Carbon Ratio Required for Mixed-Gas-Cycle Reformer Operating on Diesel Fuel

similar number of cells as the Army 1.5-kW methanol power plant and established reformer operation on diesel fuel at a steam to carbon ratio of between 1.4 and 1.9.

This same procedure was repeated for methanol fuel. The oxygen to carbon ratio to the reformer was again held fixed at 0.37 moles of oxygen per mole of carbon while the steam to carbon ratio was varied. Figure 2-5 compares the methanol and diesel results. It shows that a power plant could be operated on either fuel at 1.5 moles of steam per mole of fuel using a fuel cell stack consisting of 80 cells. The results of these thermodynamic studies defined the test conditions to be run, and the steam-to-carbon ratio and oxygen-to-carbon ratio goals to be achieved.

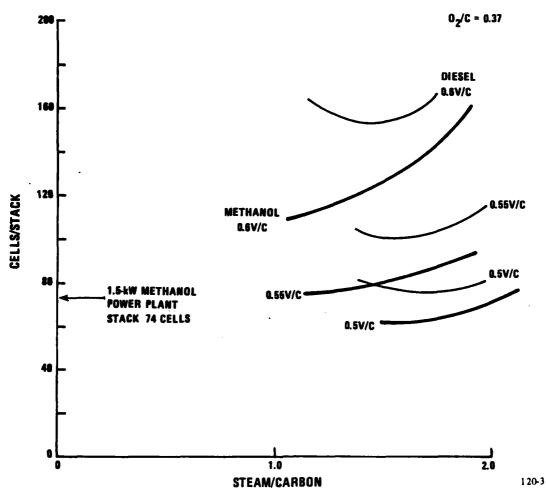


Figure 2-5. Comparison of Steam to Carbon Ratio Required with Diesel Fuel and Methanol Fuel

EXPERIMENTAL EVALUATION OF THE ADIABATIC REFORMER IN MIXED-GAS-CYCLE

All tests run during this program were conducted in a two-inch diameter bench scale reactor capable of operating with up to three pounds of diesel fuel per hour, and up to five and one half pounds of methanol per hour. The same catalyst loading used for 1412 hours in the previous adiabatic reforming study (2) was used for the 782 hours of testing in the present mixed-gas-cycle study. Three different fuels were used to determine the operating characteristics of the adiabatic reformer under mixed-gas-cycle conditions: diesel fuel, which was used as the baseline fuel, methanol and methanol contaminated with diesel fuel.

Reactor Description and Operation

The bench scale adiabatic reformer was constructed of 2-inch schedule 40 Inconel-601 pipe approximately 24-inches long. The reactor was externally insulated. It operated adiabatically in that it was heated by internal combustion of fuel and air. There was significant heat loss amounting typically to 2000 Btu/hr or about 180°F loss from the calculated adiabatic exit temperature. A schematic of the test rig is shown in Figure 2-6. Fuel was vaporized by a portion of the total steam inventory, Steam-1. The mixture was delivered at about 700°F to the nozzle section where it was mixed with air and the remaining steam, Steam-2, superheated to about 1650°F. The temperature of the final mixture before any reaction occurred, the pre-reaction temperature, could be adjusted between 900°F and 1400°F. The reactant mixture was injected into the catalyst bed where the product gas composition was sampled and the temperature measured at intervals axially down the reactor. Reactor temperatures, pressures, and gas flows were automatically monitored to shut down the reactor if pre-set operating limits were exceeded. The product samples passed through a condenser so that the composition of the dry gas was reported. In typical operation the reactor was started by feeding preheated steam and nitrogen to raise the catalyst temperature to about 1200°F. Hydrogen, air, and fuel were then added in sufficient flow to set the desired operating conditions. The hydrogen flow was set to simulate a condition in which some gas was recycled from the fuel cell anode vent. The pressure could be regulated from 10 to 30 psig.

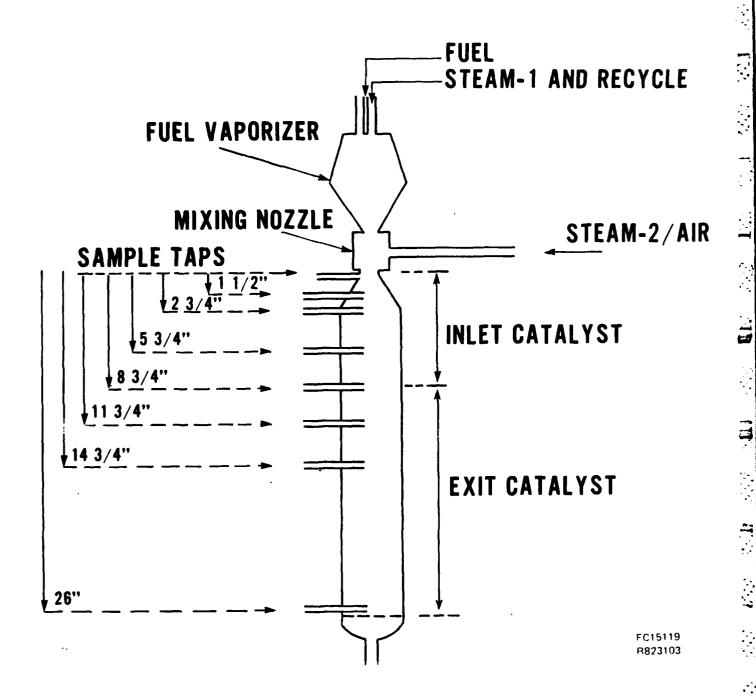


Figure 2-6. Schematic of Bench-Scale Adiabatic Reformer

A detailed diagram of the test apparatus for operation in the mixed gas cycle mode is shown in Figure 2-7.

Facility steam was superheated to 450°F before being split into Steam-1 and Steam-2. Steam flow was controlled by maintaining a predetermined pressure drop across calibrated orifices. Because previous experience had required that the fuel be vaporized by being mixed with hot gas in a spray nozzle, simulated anode gas, H₂, CO₂ and N₂ were mixed with a small amount of steam, Steam-1, and preheated to between 700°F and 925°F to supply the heat of vaporization for the fuel. To simulate cathode exhaust, and maintain a thermal balance between heat exchanger tubes of a gas fired reactant preheater, air was added to Steam-2 upstream of the reactant preheater, and cathode N₂ added through a separate tube of the reactant preheater. Both tubes were welded together at the exit of the preheater so that the gases were mixed to simulate cathode air prior to entering the fuel-air mixing The reactant preheater consisted of a down-fired methane burner with two ten foot coils of 0.5-inch 1-601 tubing and one two foot section of 3/8 inch 1-601 tubing immersed in a bed of one-half inch ceramic spheres. Simulated cathode air temperatures could be set very accurately by varying the methane flow rate to the burner. This was the primary method used to vary the pre-reaction temperature.

The mixed-gas-cycle used the same catalyst bed to reform the fuel to hydrogen rich gas, and the same gas cleanup system to eliminate sulfur from the effluent. The chloride guard had been installed to insure removal of any halogens which might have been present in coal liquids run in a previous study, and was not required during this program. Figure 2-7 also shows the placement of the three catalysts in the reactor. The inlet section was filled with PSD3018, a carbon tolerant metal oxide on a refractory support, while the exit section contained PSD2001, a high activity nickel catalyst on a refractory support. A small section between the two catalysts contained HGC1030, a commercial nickel reforming catalyst which had lower activity, but greater high temperature stability than PSD2001.

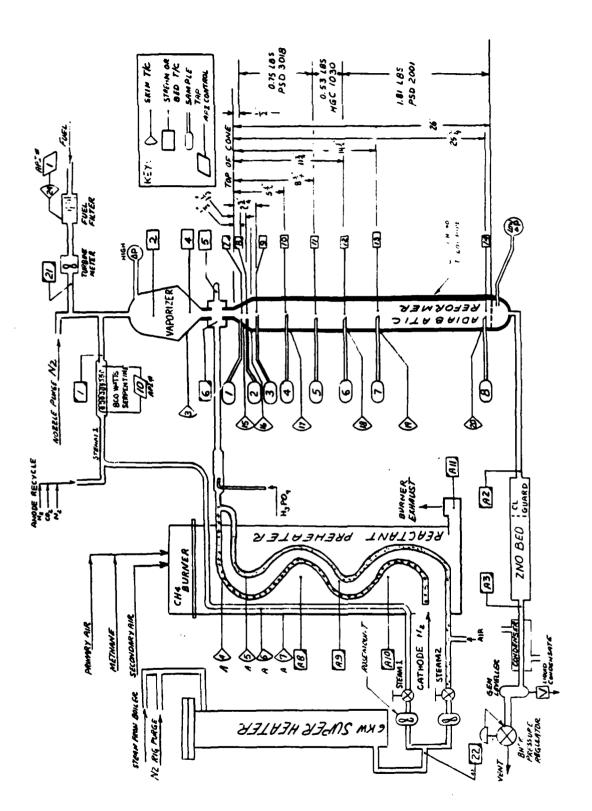


Figure 2-7. Schematic of Mixed-Gas-Cycle Test Apparatus

Gas Analysis and Sampling

Eight sample taps were located along the length of the reactor as shown in Figure 2-7. The gas from these taps would be dried and routinely analyzed with a gas chromatograph to determine the fuel conversion along the length of the reactor. A dual column arrangement consisting of Poropak®R to separate carbon dioxide, ethane, ethylene and propylene, and molecular sieve to separate hydrogen, carbon monoxide and methane was used in a Hewlett Packard Model 5700A gas chromatograph to measure the gas concentration with a thermal conductivity detector. A Shimadzu Model CE1B Data Processor was used to obtain gas concentrations from the chromatograph peaks. An example of the form in which the data for each test point was tabulated is shown in Figure 2-8.

The gas chromatograph routinely used does not measure any heavy hydrocarbons which might still be present in the reactor. The calculation used to determine fuel conversion assumes that all of the heavy hydrocarbons have been gasified to carbon oxides and hydrocarbons of C-3 or lighter. A Hewlett Packard Model 5720 flame ionization detector was used to measure the total hydrocarbon effluent from the reformer to insure the absence of heavy hydrocarbons exiting the reformer. The chromatograph column was eliminated to avoid the loss of any component through adsorption on the column material. A hot sample was fed continuously to the detector until a steady reading was observed, and then the total hydrocarbon concentration obtained in this manner was compared with the methane concentration obtained by individual component analysis. The results from both gas analysis techniques are compared in Figure 2-9. The similar values reported by both methods of analysis verified the assumption that no significant concentration of liquid hydrocarbons was present in the reformer effluent over the range of conditions tested.

The reformer effluent was also analyzed for ammonia. The analysis was performed on-line with an HNU photoionization detector Model P101. All sample lines to the detector were maintained hot in order to eliminate condensation and ammonia removal upstream of the detector. Ammonia separation was initially attempted in a six feet long by 1/8 inch diameter Poropak Q chromatograph column coated with

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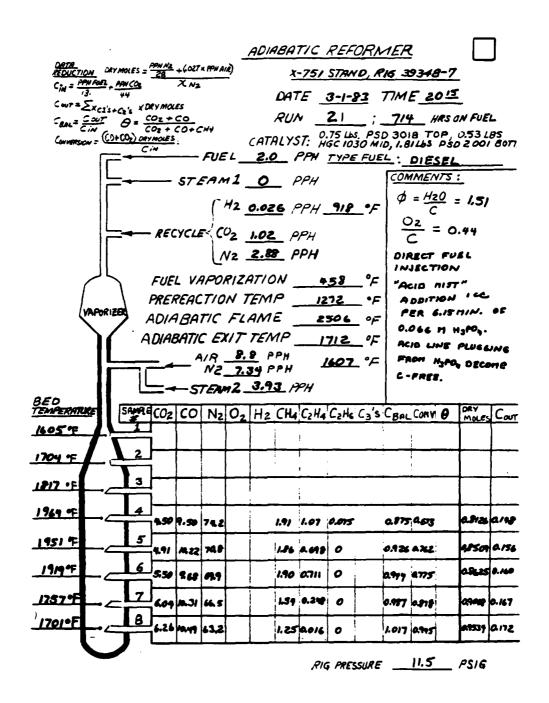


Figure 2-8. Typical Reduced Data Point Tabulation

EXIT TEMP.	CH ₄ EXIT CONCENTRATION BY GAS CHROMATOGRAPH	TOTAL HYDROCARBONS IN HOT REFORMER EXIT GAS BY F.I.D.
1677 ⁰ F	1.03%	0.8%
1735 ⁰ F	0.78%	0.7%

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Figure 2-9. Reformer Effluent Methane Concentration Measured by Gas Chromatography Compared with Total Hydrocarbon Measured by Flame Ionization Detector

15% PIE. In order to eliminate trace quantities of ethylene from interfering with the ammonia analysis, the column length had to be increased to 12 feet.

Test Facility Calibration

resting was started with diesel fuel at conventional adiabatic reforming conditions in order to establish that the reformer performance was similar to previous tests (2). It confirmed that the catalyst performance was essentially unchanged and that the new batch of diesel fuel, apparently similar in analysis, also resulted in similar conversion and carbon boundary.

Point 1C (run 21) was tested at exactly the same conditions as point 49 of the previous study (run 20). Reduced data from both points is shown in Figure 2-10. Run conditions are shown in the top part of this figure, and gas analysis, temperatures and reduced data are shown next to the reformer sample tap with which they are associated. The results of both tests agree very closely. Conversion to carbon oxides, θ , defined as $(CO + CO_2)/(CO + CO_2 + CH_4 + 2 \times C_2H_4 + ----)$ agree within one percent; the carbon balance which is defined as the moles of gaseous carbon exiting the reformer $(CO, CO_2, CH_4, 2 \times C_2H_4, etc.)$ divided by the moles of carbon entering the reformer agree within one percent at three of the four taps analyzed; temperatures are similar, and both points are at the carbon boundary.

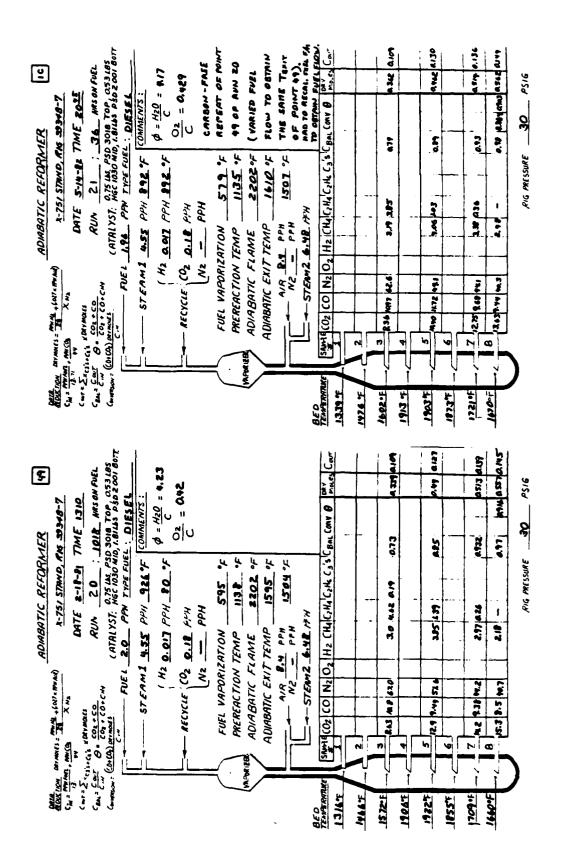


Figure 2-10. Reduced Data from Runs 20 and 21

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In an adiabatic reformer operating on heavy distillates, the prevention of carbon formation in the reformer is very important. For a given catalyst and reactant mixing nozzle configuration, carbon formation is prevented by either heating the reactants prior to their entering the reformer, or by adding excess air. The reformer carbon boundary is determined by the following procedure. Air flow to the reformer is reduced until carbon is formed as indicated by an increase in the pressure drop across the reformer catalyst bed. After a definite increase in pressure drop is observed, air flow to the reformer is incrementally increased until the pressure drop starts to decrease. This process is repeated over a range of reactant preheat (pre-reaction) temperatures until a carbon boundary is defined as shown in Figure 2-11.

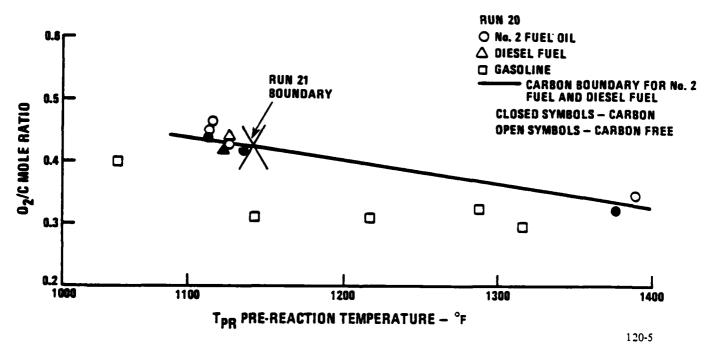


Figure 2-11. Comparison of Carbon Boundary from Runs 20 and 21

The solid line is the carbon boundary which was defined for No. 2 fuel oil and diesel fuel during run 20. Operating above the line resulted in carbon-free operation whereas operating below the line resulted in carbon formation in the reformer. Gasoline, a light distillate fuel did not form carbon even at very low oxygen to fuel carbon mole ratios. The calibration point for run 21 duplicated run 20 exactly, and is shown as a large X falling right on the old carbon boundary.

Fuel conversion was slightly lower in the new run, and is shown plotted in Figure 2-12. Because fuel conversion is 1.3% lower, a slight decrease in fuel conversion during mixed-gas-cycle operation would be the result of a slight decrease in the catalyst activity, not in the different mode of operation.

Carbon Boundary During Mixed-Gas-Cycle-Operation

Operating in a mixed-gas-cycle mode results in operating at a very low steam to carbon ratio, and with a reactant stream greatly diluted with nitrogen. Conventional steam reformers require a minimum steam to carbon ratio of about 2.5 to avoid carbon formation. Adiabatic reformers have the potential for operating at lower steam to carbon ratios than conventional reformers because of air addition and the higher operating temperatures. Both air and increased temperature enhance carbon gasification, and the added air also results in some steam production from the combustion of hydrogen.

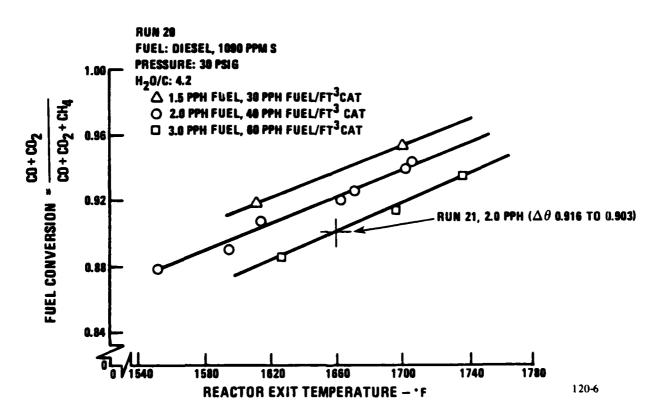


Figure 2-12. Fuel Conversion vs. Reactor Exit Temperature

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Previous tests had run at 30 psig; however to conserve compressor parasite power requirements, a power plant would run as close to ambient pressure as possible. The original test hardware was designed to operate at high pressure, and back pressure through the exhaust system did not allow the reactor to operate much below 30 psig until modifications were made. Initial mixed gas cycle tests, run at 30 psig and a steam to carbon ratio of 1.91, resulted in a carbon boundary which certainly would have been unacceptable to a conventional power plant operating with an adiabatic reformer. When pressure was lowered to 12 psig, the minimum possible with the available test hardware, the carbor boundary improved significantly as shown in Figure 2-13. Decreasing the steam to carbon ratio to 1.5, the lower limit required during mixed-gas-cycle operation, resulted in a slight, but still acceptable increase in the carbon boundary as shown in Figure 2-14.

Fuel Conversion During Mixed Gas Cycle Operation

Diesel fuel was tested at 1.5 and 2.0 pounds per hour over a range of exit temperatures in the mixed-gas-cycle mode. Experience has shown (1) that at a given fuel flow rate, fuel conversion can be correlated with catalyst exit temperature regardless of how that exit temperature is achieved. Fuel conversion is shown over a catalyst exit temperature range of from 1620 to 1780°F in Figure 2-15. It shows fuel conversion following the expected trend of increasing with increasing temperature and decreasing fuel flow.

A more convenient way to describe the fuel flow rate to a reactor is in units of space velocity defined as the specific fuel flow rate divided by the reactor (or catalyst) weight or volume. Space velocity is convenient, especially for design purposes, since it defines the reactor size when the fuel flow rate is known.

Previous studies (1) have shown that a fuel conversion of 98.2% is required to obtain high power plant efficiencies with an adiabatic reformer. A space velocity of 12 pounds of fuel per hour per cubic foot of catalyst is required to obtain this conversion at a 1700°F catalyst exit temperature with a conventional adiabatic reformer. A simple cross-plot of the data from Figure 2-15 compares conversion

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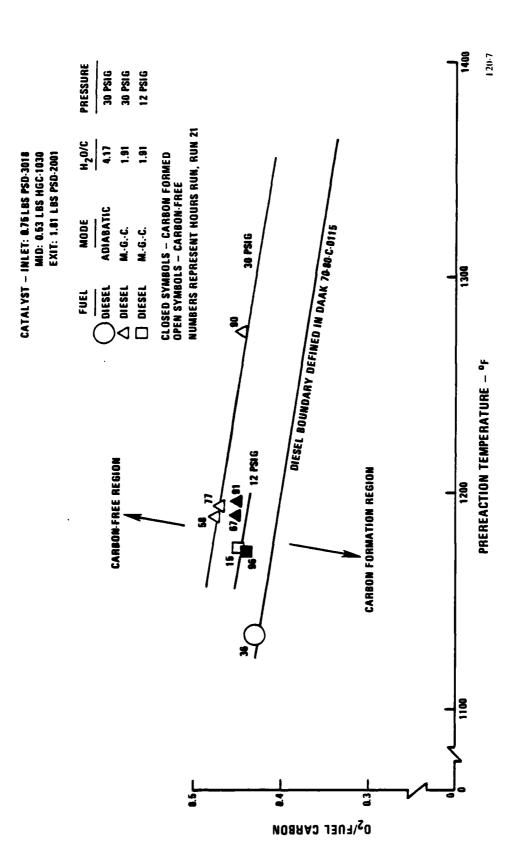


Figure 2-13. Effect of Pressure on the Carbon Boundary

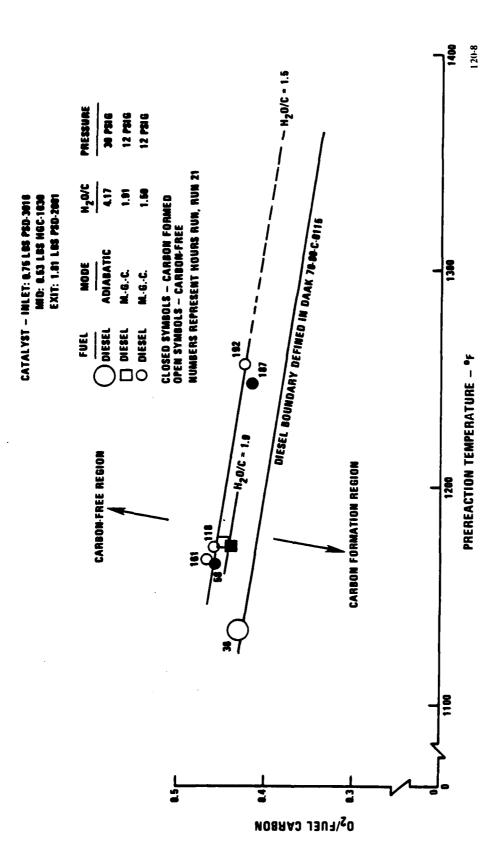


Figure 2-14. Effect of H₂O/Carbon on Carbon Boundary

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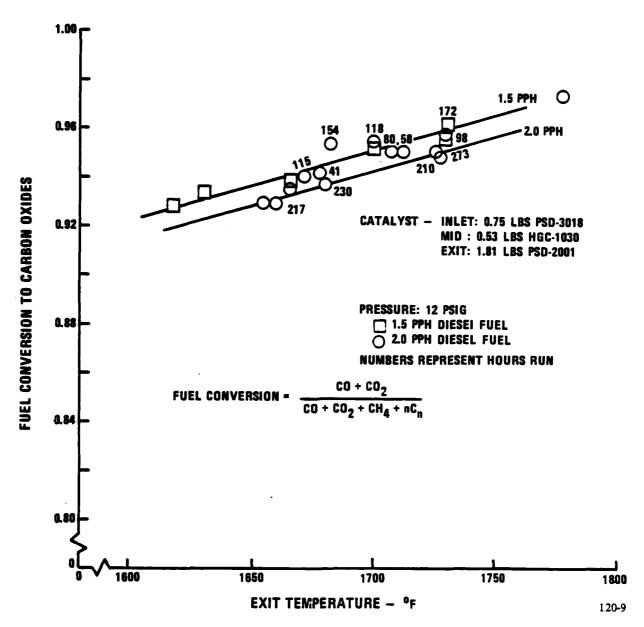


Figure 2-15. Fuel Conversion in Mixed-Gas-Cycle vs. Reactor Exit Temperature

results obtained both with conventional adiabatic reforming and with adiabatic reforming in the mixed-gas-cycle mode. These results, plotted in Figure 2-16, show that conversion is similar during both modes of operation, and that the conversion obtained during mixed-gas-cycle operation might reasonably be extrapolated to show that the desired fuel conversion can be achieved.



• MIXED-GAS-CYCLE, H₂O/C = 1.5, DIESEL FUEL, 12 PSIG

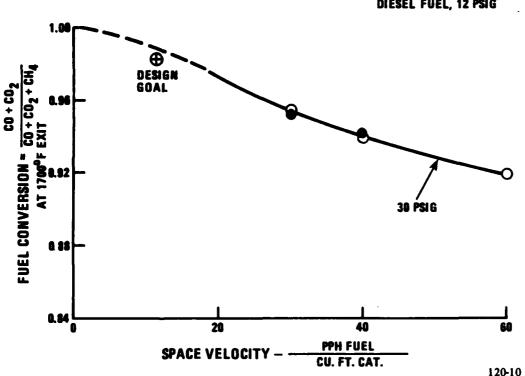


Figure 2-16. Fuel Conversion in Mixed-Gas-Cycle Mode and Conventional Adiabatic Reforming

Both pure and contaminated methanol were tested. Diesel fuel was used to contaminate the methanol in order to simulate contamination which might occur in a field situation where a fuel container would be used interchangeably between methanol and diesel fuels. The 5% contamination is the greatest quantity of diesel which could be dissolved in 100°F methanol. Because methanol is a partially oxygenated fuel with a much lower heating value than diesel fuel, a higher methanol fuel flow rate is required to produce the same power rating as with diesel fuel. Equivalent methanol flows were 74% greater than with diesel fuel, so 3.48 pounds per hour of methanol is equivalent to, and compared with 2.0 pounds per hour of diesel fuel.

As might be expected, clean methanol containing less than one part per million of sulfur, was very easy to reform. Even at the high space velocities used during this series of tests, 99% fuel conversion was achieved at about 1500°F. Five percent contamination with diesel fuel resulted in about two percent loss in conversion compared with pure methanol at identical conditions. These results are compared in Figure 2-17 with the much lower conversion of diesel fuel.

CATALYST: INLET 0.75 LBS PSD-3018
MID 0.53 LBS PSD-1030
EXIT 1.81 LBS PSD-2001

FUEL:

○ METHANOL: 3.48 PPH (EQUIV. TO 2.0 PPH DIESEL)
△ METHANOL: 5.22 PPH (EQUIV. TO 3.0 PPH DIESEL)
SOLID SYMBOLS: METHANOL CONTAMINATED
WITH 5% DIESEL

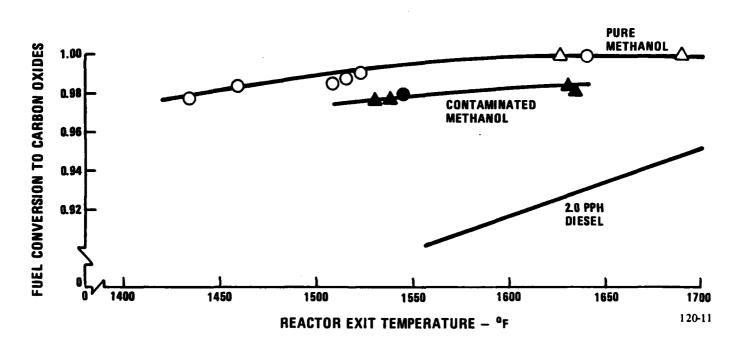


Figure 2-17. Methanol Fuel Conversion

Product Gas Characterization for Ammonia Content

Nitrogen present in the adiabatic reformer has the potential for reacting with hydrogen to form ammonia in the reaction:

$$N_2 + 3H_2 \stackrel{?}{=} 2NH_3$$

It was not certain to what extent ammonia formed; however any ammonia which reaches the phosphoric acid fuel cell reacts with the acid to form solid ammonium phosphate which constricts the cell flow passages.

$$H_3PO_4 + 3NH_3 \stackrel{\rightarrow}{\downarrow} (NH_4)_3 PO_4$$

Analysis of the hot reformer effluent showed a very low concentration of less than ten parts per million of ammonia present in the reformer effluent. The equilibrium value is the maximum concentration of ammonia possible in the reformer. Equilibrium values were calculated at two pressure levels: 1.7 atmospheres represents the pressure at which the test was conducted, and 1.0 atmospheres represents the expected power plant pressure. The measured ammonia concentrations of less than ten parts per million were significantly below equilibrium as shown in Figure 2-18. The cross-hatched line represents the maximum value of ammonia that could be present in the reformer effluent gas stream based on the gas chromatograph response. Although the quantity of ammonia formed is very small, it still requires that some type of scrubbing device be used in a power plant to lower the ammonia effluent to less than one part per million in order to eliminate the potential for plugging of anode passages.

Direct Fuel Injection Nozzle Evaluation

A significant step in the development of a mixed-gas-cycle power plant is the direct injection of fuel into the reformer. Prevaporization of the fuel prior to injection into the reformer was a convenient test vehicle assuring complete fuel

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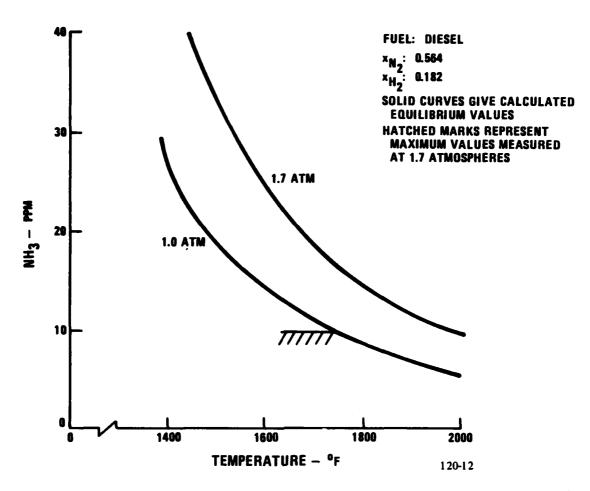


Figure 2-18. Ammonia Concentration on Mixed-Gas-Cycle Reformer Effluent

vaporization and mixing with the other reactants; however the fuel nozzle used to prevaporize the fuel by mixing would result in significant penalties for a power plant.

A fuel injector was designed which allowed fuel to be injected directly into the hot gas stream above the catalyst without the requirement for prevaporization. The design allowed the injector to be used with minor modifications to the rig while maintaining the vaporizer intact. Figure 2-19 shows the general placement of the injector relative to the vaporizer, mixer and catalyst bed. Fuel was deposited on a flat plate located at the end of the injector and placed in the mixing section of the reactor.

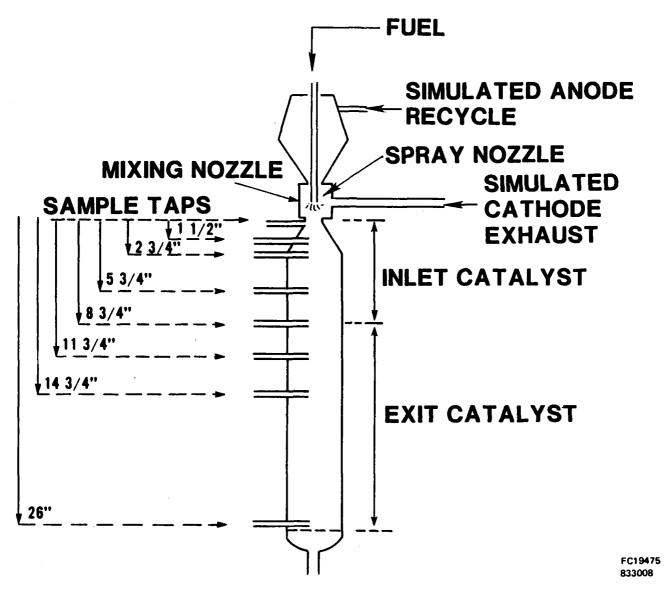


Figure 2-19. Placement of Direct Fuel Injection Nozzle

The details of the injector are shown in Figure 2-20. A hypodermic tube used to inject the fuel was supported in a series of one-eighth and one-quarter inch tubes. A flat splash plate was supported with two 1/16 inch weld wires 1.5 inches below the hypodermic tube exit. This entire assembly was centered in a 3/4 inch tube attached to a 3/4 inch A/N fitting with two 1/16 inch weld wires. The flexibility of the weld wires allowed the injector assembly to be centered in the mixing section even if the fitting at the top of the vaporizer and the mixing section at the

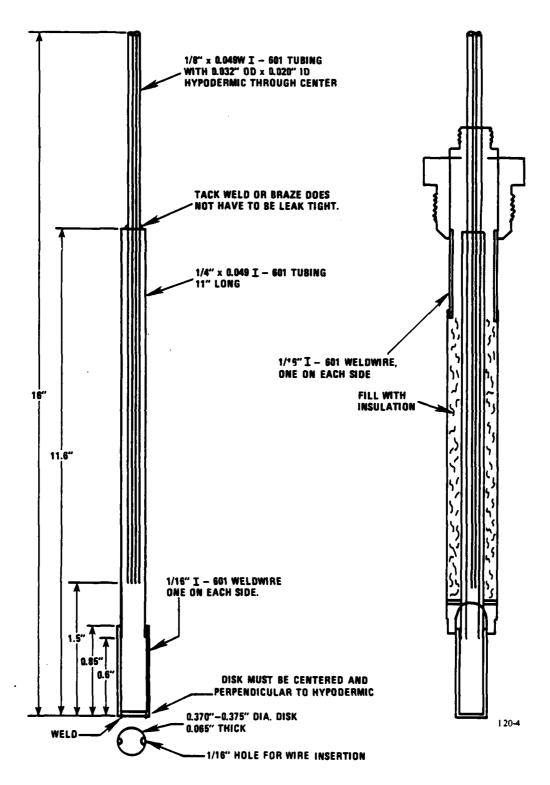


Figure 2-20. Fuel Injector Construction

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bottom of the vaporizer were not concentric. A 1/4 inch compression type fitting was welded to the top of the 3/4 inch fitting, and a 1/4 inch to 1/8 inch teflon reducing ferrule was used to make a gas tight seal while still allowing the center assembly to be moved up and down along the axis of the mixing section. Insulation was packed between the inner assembly and the 3/4 inch tube in order to insure that the fuel remained cool until injected into the mixing section. A picture of the completed fuel injector is shown in Figure 2-21.

The greatest concern at the start of this test was that direct fuel injection would result in incomplete fuel vaporization and would not provide adequate mixing of the fuel with the steam and air. Operation at a very high oxygen to carbon ratio would then be required to prevent carbon formation. Tests with the direct fuel injection nozzle resulted in a carbon boundary which was identical to the carbon boundary with prevaporized fuel as shown in Figure 2-22. The use of direct fuel injection was so successful, that it was continued during the exhaust impurities testing.



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Figure 2-21. Completed Fuel Nozzle

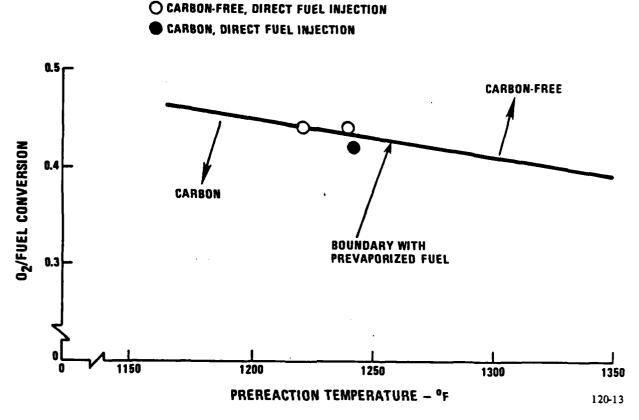


Figure 2-22. Carbon Boundary with Direct Fuel Injection

Effect of Fuel Cell Exhaust Impurities on Reformer Catalyst

In the mixed-gas-cycle adiabatic reformer, air for the combustion process comes from the cathode exhaust. As a result, the air contains phosphoric acid vapors from the cell. The effect of acid carryover on the reactor performance and catalyst was unknown. If the reactor or catalyst were affected by the acid carryover, carbon tolerance and conversion would be reduced.

A test was run to determine the effect of phosphoric acid addition on the performance of the cell. A Milton Roy positive displacement pump was used to pump 20 cc/hr of 0.066 molar phosphoric acid solution into the reformer for 120 hours. To insure that the acid would be volatilized and transported to the reformer, rather than becoming concentrated at some point upstream of the catalyst bed, the acid

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was pumped into the 1600°F-1650°F simulated cathode exhaust stream just upstream of the reformer, as shown in Figure 2-7. The amount of acid injected into the reformer represented the effluent from a 400°F cathode operating at a pressure of one atmosphere.

The carbon boundary shown in Figure 2-23 remained unchanged during acid addition to the reformer. Figure 2-24 shows the fuel conversion throughout the direct fuel injection testing, and it shows stable fuel conversion throughout the test, including during the period when acid was being added to the reformer.

During that time, two liquid condensate samples were taken. They were clear and odorless, indicating complete liquid fuel conversion, and when analyzed, they were free of phosphate ions. An analysis of the zinc oxide sulfur scrubber located

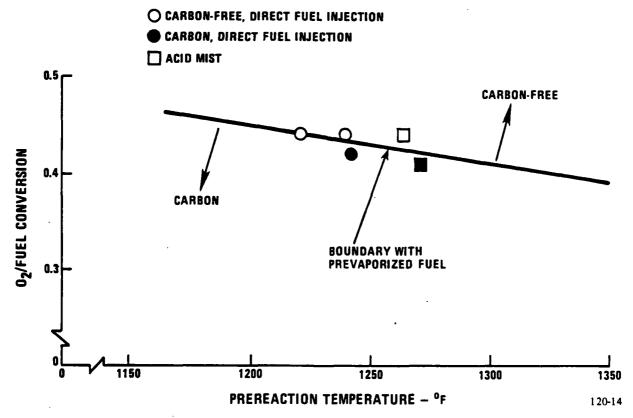


Figure 2-23. Carbon Boundary During Acid "Mist" Addition

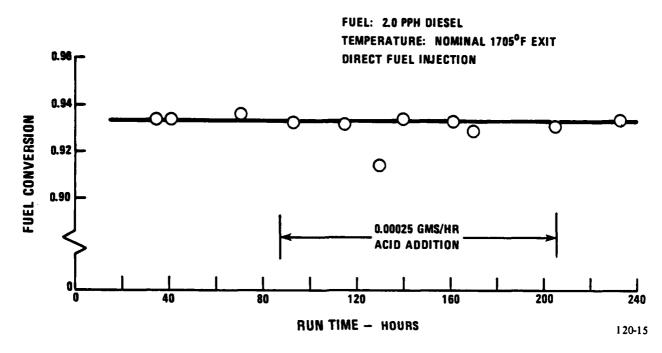


Figure 2-24. Conversion During Acid Addition

downstream of the reformer, and of the reform catalyst, indicated that the phosphoric acid had passed through the reformer and reacted with or deposited on the zinc oxide. Most probably, zinc phosphate was formed.

Post Test Catalyst Evaluation

At the end of the program, the reactor was cut open to examine the catalyst. The overall catalyst level had dropped 4-1/4 inches. The inlet metal oxide catalyst extrusions looked normal, but had shrunk 9.5% based on the average diameter of fifteen measured pellets selected at random. The mid catalyst, which was a commercial secondary reforming catalyst did not change in size. The exit high activity nickel catalyst not only had shrunk 10.5%, but it had become significantly weaker and many of the pellets had broken or powdered. A photo showing the broken pellets and comparing the fresh and used exit catalyst is shown in Figure 2-25. Previous tests run at PSD had shown that this catalyst would be weakened from operating above 1700°F and some strengthening agents were added to the basic catalyst for these tests; however, it obviously still needs improving in order

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to survive high temperature adiabatic reforming conditions and the rough handling of a military power plant. No effect of acid or exhaust impurities on the catalyst was observed.

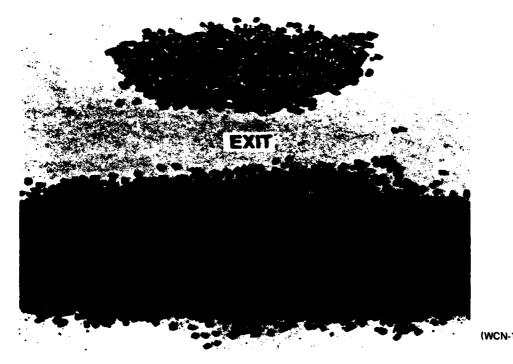


Figure 2-25. Comparison of Fresh (Above) and Used (Below) High Activity Exit Catalyst

PRELIMINARY COMPONENT DEFINITION AND SYSTEMS ANALYSIS

A system study was conducted to define a system configuration for a mixed-gascycle power plant using diesel fuel. With the defined system configuration, efficiency, weight and volume were estimated for a 1.5-kW power plant and then these same characteristics were extrapolated to power plants with ratings of up to 100-kW. Key technology areas were identified and potential system improvements recommended.

Fuel processor operating conditions were based largely on the results of the present experimental test program, while the fuel cell stack was maintained as close to the present 1.5-kW air-cooled methanol power plant cell design as possible. All of

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the power plant characteristics were calculated based on the 1.5 to 100-kW power rating of the power plant; part load, transient response and start-up requirements were considered to be outside of the scope of this study.

The key assumptions made for the system study are shown in Table 2-3. A typical diesel fuel was used; however in order to insure that the zinc oxide sulfur removal bed was of adequate size, a "worst case" sulfur content of 5000 parts per million was assumed. This sulfur level assumption results in a large zinc oxide reactor bed but produces a very conservative bed size.

The fuel cell stack was based as much as possible on the present 1.5-kW Army methanol power plant. Notable differences include anode gas cooling to maintain the cell temperature, and operation with 70% oxygen utilization in the cathode. Other key assumptions are based on PSD fuel cell power plant experience, and in the case of pump and blower efficiencies, based on commercial hardware specifications.

A schematic of the power plant as defined in the system study is shown in Figure 2-26. It is similar to the generalized schematic shown in Figure 2-2; however only those components required for operation of the power plant are shown in Figure 2-26. Four blowers and four heat exchangers are required for power plant operation. Air is used not only in the cathode and burner, but also for cooling of the anode recycle and the shift converter influent. Gases to the adiabatic reformer must be raised to a high temperature in order to obtain high temperatures and a high fuel conversion in the reformer. Gases to the zinc oxide bed and shift converter (low temperature S/C) must be cooled to about 400°F in order to maintain adequate equilibrium conditions:

in the zinc oxide bed:

$$ZnO + H_2S \stackrel{?}{\downarrow} ZnS + H_2O$$

and in the shift converter:

$$CO + H_2O \stackrel{\rightarrow}{\leftarrow} CO_2 + H_2$$

TABLE 2-3. 1.5 KILOWATT MIXED GAS CYCLE FUEL CELL SYSTEM KEY ASSUMPTIONS

Fuel:

Diesel Oil - CH1.8

Higher heating value - 19500 Btu/lb

5000 ppmw sulfur

Fuel Cell: Based on the Army 1.5 Methanol Fuel Cell

Cooled by recycling anode fuel gas

1 atm operating pressure

340°F average cell temperature

0.283 ft² active area/cell

90 watts/ft² baseline cell performance

90% H₂ utilization (internal 23%)

70% O₂ utilization

Fuel processing: Based on experimental tests of the adiabatic

reactor for the mixed-gas-cycle

1600°F exhaust temperature

0.4 moles O₂ per atom of fuel carbon

1.82 moles H₂O per atom of fuel carbon

Space velocity = $12 \# fuel/(hr-ft^3 catalyst)$

Desulfurization: Disposable ZnO beds

Sulfur slip = 0.1 ppm (Based on wet gas volume)

Ammonia scrubber: H₃PO₄ acid-charcoal bed

NH₃ leaving reactor < 10 ppmv

 NH_3 to fuel cell < 0.5 ppm \vee

Shift Converter: Temperature catalyst

CO slip = 1% (Dry Gas)

Inverter efficiency = 85% (1.5-kW size)

Pumps-blower efficiencies scaled from commercial hardware specs.

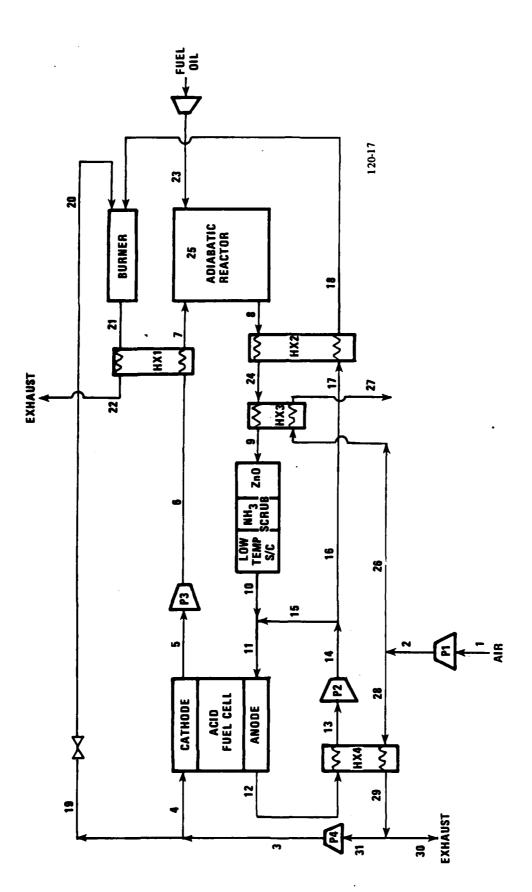


Figure 2-26. Power Plant System Schematic.

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Furthermore, the shift catalyst must be maintained below about 550°F to prevent activity loss by sintering. The fuel cell is maintained at the relatively low temperature of 340°F in order to prevent drying out from loss of phosphoric acid electrolyte at the low operating pressures of this power plant.

Power plant efficiency was obtained by accounting for the parasite power requirement and efficiency of each component in the system. Pump and inverter efficiencies are shown as an example in Figure 2-27. As might be expected, increasing the size of these components, as is required with increasingly larger power-plants, results in increased efficiency for each component. The overall power plant efficiency after accounting for the efficiency losses from each component is shown in Figure 2-28 as a function of power plant rating.

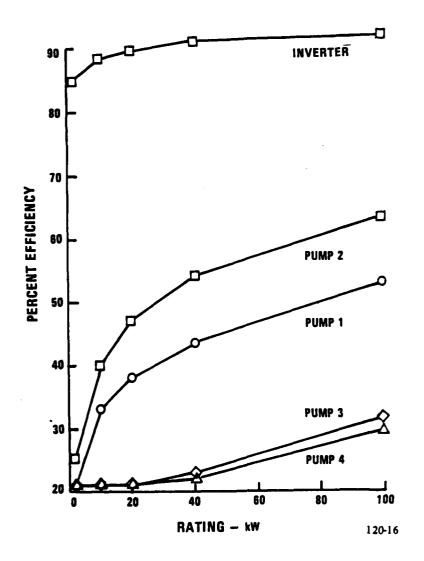


Figure 2-27.

Typical Component Efficiencies

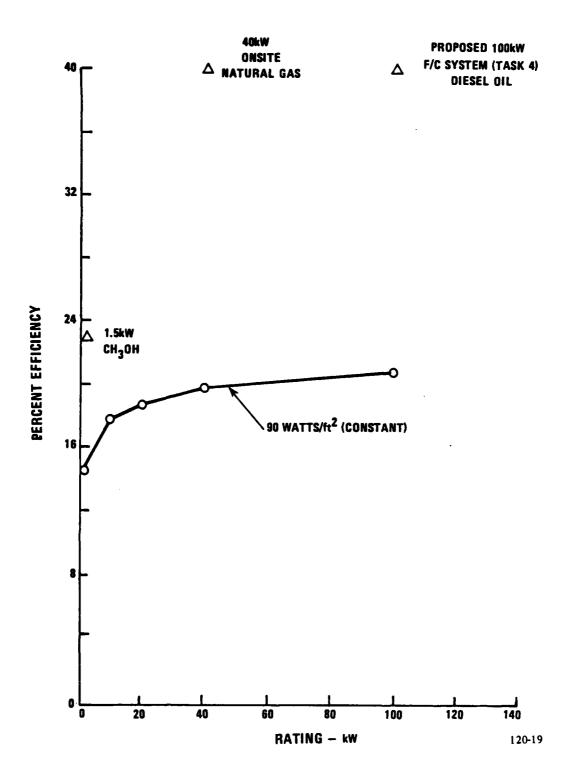


Figure 2-28. Power Plant Efficiency vs. Rating

Power plant operating characteristics were studied for five power ratings: 1.5-kW, 10-kW, 20-kW, 40-kW, and 100-kW. Several baseline power plants are shown in the figure for comparison purposes. The 1.5-kW diesel mixed-gas-cycle power plant is 63% as efficient as the 1.5-kW Army methanol power plant. Efficiency increases with power plant rating, and at 100-kW, the efficiency of the mixed-gas-cycle power plant is 95% of the 1.5-kW Army methanol power plant. Highest efficiencies are obtained with the 40-kW on-site natural gas power plant, and the proposed 100-kW cyclic fuel cell system operating on diesel fuel and described in detail in the Task 4 section of this report.

Both specific size and weight are dramatically decreased as the power plant rating is increased from 1.5-kW to 10-kW, as shown in Figures 2-29 and 2-30. Although the 40-kW on-site natural gas power plant is smaller and lighter than the equivalent rated diesel fueled mixed gas cycle power plant, Figures 2-29 and 2-30 compare these power plants and show that both size and weight of the mixed gas cycle power plant approach the 40-kW on-site power plant. The design for the present study was based on rated power, and did not include provisions for overload which are included in the on-site design.

A major consideration in any fuel cell power plant operating on high sulfur fuel is the sulfur removal system. This study assumes that the fuel contains 5000 parts per million of sulfur by weight, and that all of the sulfur is removed with a discardable zinc oxide cartridge. In order to minimize the size impact of the sulfur scrubber, a changeout period of 30 days is assumed in the design. Even with such frequent changes, the sulfur scrubber accounts for about 20% of the power plant volume. Because a large percentage of the zinc oxide bed is used to remove the final traces of sulfur from the gas stream, increasing the changeout period improves the effectiveness of the zinc oxide. Figure 2-31 shows the impact of the zinc oxide weight on its replacement period. Increasing the replacement period by a factor of three, from 30 days to 90 days, increases the required zinc oxide weight by only 34%. Although a 90 day replacement period (2160 hours) might be more desireable from an operational point of view, it would result in a significant impact on power plant size. Details of this study were presented to MERADCOM personnel at Ft. Belvoir on March 22, 1983. A copy of this presentation is included in this report as Appendix C.

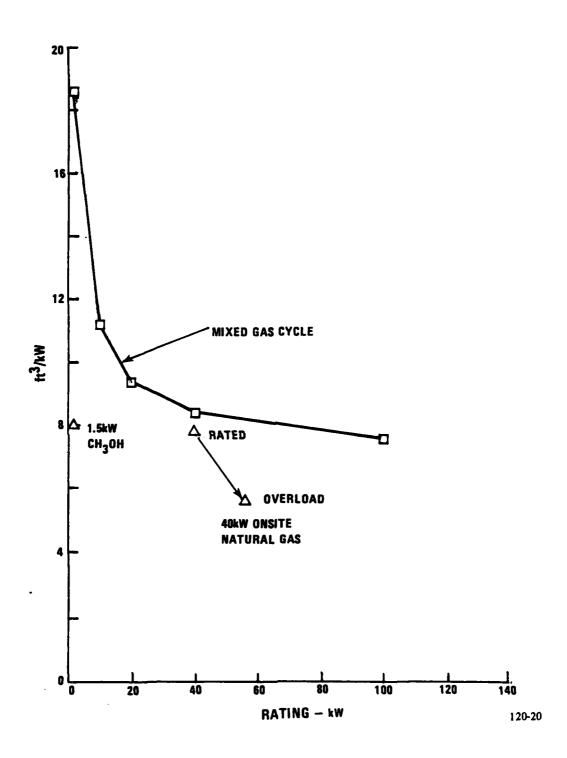


Figure 2-29. Power Plant Specific Volume vs. Rating

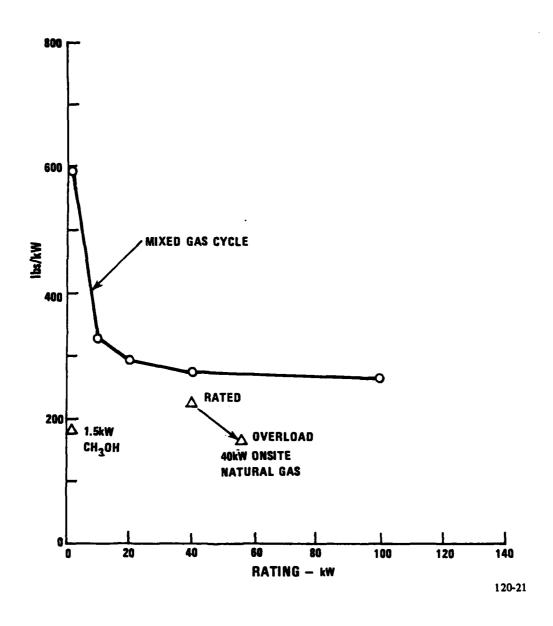


Figure 2-30. Power Plant Specific Weight vs. Rating

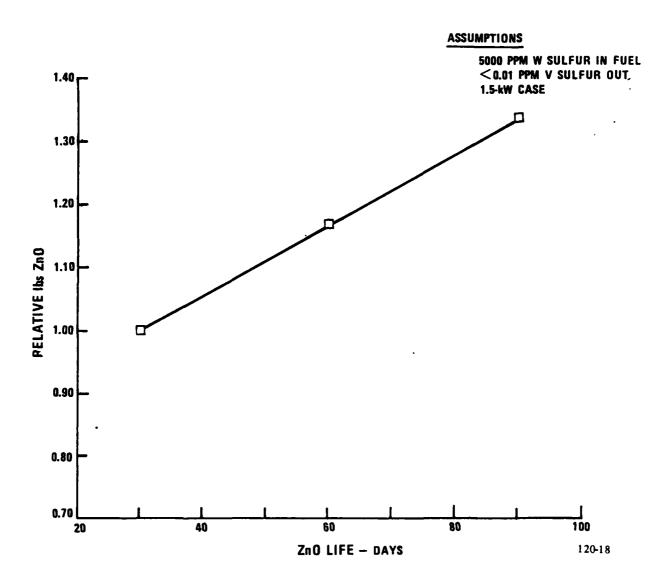


Figure 2-31. Impact of ZnO Weight on Replacement Period

DISCUSSION OF THE MIXED-GAS-CYCLE POWER PLANT

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Tests and system studies have demonstrated that an adiabatic reformer operating in a fuel cell power plant in the mixed-gas-cycle mode has the potential to provide multi-fuel capability for military fuel cell power plants. Operating conditions were selected for operating both methanol and diesel fuels in a mixed-gas-cycle power-plant which would require no water addition, and have no water recovery system which might result in potential power plant freeze problems. A thermodynamic

system match was obtained for both fuels. They were then evaluated by tests, and the reformer found to be capable of operating within the window defined by the system for both fuels.

Although operation with diesel fuel in the mixed-gas-cycle mode requires more air addition to the reformer than for conventional adiabatic reforming to prevent carbon formation, the decreased mixed-gas-cycle power plant operating pressure lowers the air requirement, and results in a carbon boundary which matches power plant system requirements for a steam to carbon ratio of 1.82. See Figures 2-13 and 2-14. Test hardware contraints limited the lower operating pressure to about 12 psig, whereas the power plant would operate at ambient pressure, approximately 0 psig. This lowered power plant pressure would result in some safety factor in the carbon boundary during power plant operation. Carbon formation would not be considered a problem for methanol.

Diesel fuel conversion was the same for adiabatic reforming in the conventional mode or in the mixed-gas-cycle mode. Conversion extrapolates to the system requirement for both modes. See Figure 2-16. Conversion with methanol is significantly greater than with diesel fuel. See Figure 2-17. This is an expected result, since one of the most critical factors affecting fuel conversion is sulfur poisoning of the reform catalyst. Methanol contains less than one part per million of sulfur, whereas diesel fuel contains more than 1000 parts per million of sulfur. Analysis of both the hot gas effluent, and the water condensate from the reformer showed complete fuel conversion in the reformer. No unreacted diesel fuel or polymerized hydrocarbons were detected in the reformer exhaust.

Less than ten parts per million of ammonia was detected in the reformer effluent. Although it is a very small quantity, it must be removed to prevent blockage of the anode gas passages in the fuel cell from solid ammonium phosphate. An ammonia scrubber consisting of charcoal impregnated with phosphoric acid was projected for power plant use in the system study. It was only 1.0% to 1.5% of the power plant size depending on the power plant rating.

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Acid mist did not affect operation of the reformer. Neither fuel conversion nor carbon boundary appeared to be changed by acid addition. See Figures 2-23 and 2-24. Post test analysis indicated that acid passed through the reformer and was captured in the zinc oxide reactor; however in a power plant, there is some concern that acid might result in corrosion to some component between the cathode exit and the zinc oxide sulfur scrubber.

Sulfur removal with a high sulfur fuel such as diesel requires a large quantity of zinc oxide. It is a workable solution to the use of sulfur containing fuel; however the quantity of zinc oxide required to insure sulfur-free hydrogen gas to the shift converter and to the anode has a significant impact on the power plant size. Zinc oxide accounts for over twenty percent of the power plant volume. See Figure 2-31. In order to insure complete sulfur removal from the reformed gas, the sulfur scrubber must be sized to account for a worst case fuel. In this case, it was assumed that 5000 parts per million of sulfur would be the maximum quantity of sulfur found in diesel fuel.

Two potential solutions exist for minimizing the impact of sulfur removal. The most readily available solution is to use sulfur-free methanol as the fuel. This would not only eliminate the requirement for sulfur removal, but would also increase the efficiency of the power plant. A second solution would be to use a regenerable sulfur scrubber to remove the bulk of the sulfur, followed by a small zinc oxide bed for "polishing". It would not be reasonable to use an industrial type of liquid sulfur scrubber such as a packed tower with an amine scrubbing solution for a power plant. A more reasonable solution for a mobile power plant would be to use a solid regenerable sulfur scrubber. Although some work has been performed on solid regenerable sulfur scrubbers, none are developed to date. The use of methanol fuel is certainly a much simpler solution.

A power plant would require the fuel to be directly injected into the reformer. Previous tests had been run with prevaporized fuel to insure complete fuel vaporization and complete mixing with steam and air. This was convenient for test purposes, but not practical for power plant operation. Tests run during this

study have demonstrated that direct fuel injection is possible. A fuel injector was designed (see Figures 2-19, 2-20, and 2-21) and successfully tested. It resulted in no change in reformer performance. See Figure 2-22.

A system was defined, and an estimate made of the fuel cell stack and reformer size for various power levels for a power plant operating with diesel fuel. A 1.5-kW power plant is very large. Increasing the size to 10-kW reduced the specific size and weight significantly. See Figures 2-29 and 2-30.

RECOMMENDATIONS

Further testing as well as systems work is required to reduce the mixed-gas-cycle power plant to practice. Because much of the heat required for adiabatic reforming is generated inside of the reformer, raising the reformer temperature to start-up conditions without oxidizing the reformer catalyst will probably require some type of partial oxidation with a spark plug or glow plug inside of the reformer. Testing will be required to demonstrate a reformer start-up system which does not damage the catalyst. A high activity exit catalyst is required for operation on diesel fuel. The activity of the present catalyst is adequate; however its strength at temperatures above 1700°F is not adequate. The activity of commercial secondary reformer catalyst is probably adequate for operation with methanol. Further reformer testing should be with low pressure drop hardware which can be operated at power plant conditions.

Components which must be more fully defined include the high temperature heat exchanger, HEX 1 of Figure 2-26, the anode recycle pump, and the ammonia scrubber. A more compact (regenerable) sulfur removal system should be developed for use with diesel fuel, and corrosion caused by acid vapors should be studied.

In view of the more compact size and the potential for higher efficiency with methanol fuel, a detailed system analysis should be conducted based on the use of neat methanol as the fuel for a mixed-gas-cycle power plant. Alternate system options should be evaluated and critical components more fully defined. Start-up, part power and transient operation must also be considered in order to fully define the power plant.

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REFERENCES

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- J. A. S. Bett, R. R. Lesieur, A. P. Meyer, H. J. Setzer, "Characterization of the Performance of Adiabatic Reformers Operated with Logistic Fuels", <u>Final Report</u>. U. S. Army Contract DAAK70-80-C-0115, June 1981.

SECTION 3

ANALYTICAL STUDY TO DETERMINE PREFERRED FUEL CONDITIONING PROCESS FOR REMOTE SITE MILITARY POWER PLANT

PROGRAM OBJECTIVE

On-site fuel cell power plants are designed for use with pipeline gas and for connection to either an isolated facility or operation in parallel with a utility grid. To enable this class of power plant to be used easily at remote facilities, a study was conducted to determine if operation on Air Force logistic fuels, such as diesel oil can still achieve high power plant efficiency and utility. For this study, four alternative fuel processor concepts were investigated. The four options included:

a) thermal steam reforming, b) adiabatic reforming, c) hybrid reforming, and d) cyclic reforming. The fuel processor study comparison includes overall system efficiencies determined from thermodynamic analysis, a development risk assessment, power plant capital cost, and power plant availability.

A fuel processor subsystem capable of processing diesel fuel would result from the technical data comparison.

APPLICATION REQUIREMENTS

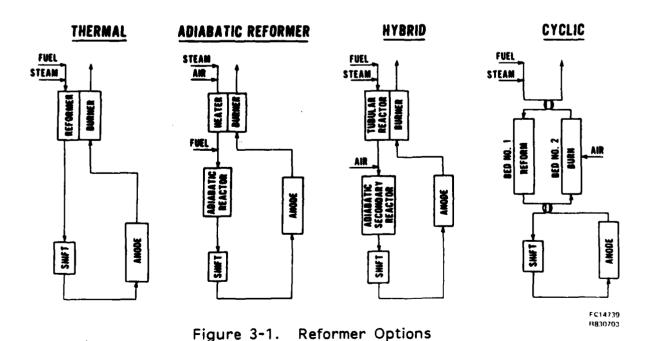
Air Force site power requirements for the study are in the 60-to-120-kW range. A nominal power level of 100 kW was selected for the power plants studied. Preferred fuel for the application is diesel. The power plant is to be sited in a remote site. This requirement sets a premium on electrical efficiency since fuel transportation costs will comprise a significant percentage of the total life cycle cost.

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FUEL PROCESSOR OPTIONS FOR LOGISTIC FUELS

Four fuel processing options were identified for handling diesel fuel for use in fuel cell power plants. Simple sketches of each of these fuel processor options are shown as Figure 3-1, respectively. They are as follows:

- a. <u>Thermal steam reforming</u>, in which fuel and steam are reacted directly over a metal oxide catalyst at a high temperature
- b. <u>Conventional Adiabatic Reforming</u>, in which air, fuel and steam are reacted directly over a nickel catalyst
- c. <u>Hybrid reforming</u>, in which a primary thermal steam reformer operating at low fuel conversion is used in combination with a secondary adiabatic reformer
- d. <u>Cyclic reforming</u>, in which two beds are cycled between a "make" mode where reformed gas is produced and a regeneration mode where the heat of reforming is provided via combustion



GENERAL ASSUMPTIONS

System operation and design is based, as much as possible, on current on-site power plant designs. These designs are evolving from the 40-kW power plants currently under field test but employ near-term technology improvements expected to be available within the next 2-4 years.

SYSTEM DESCRIPTIONS

Thermal Steam Reformer

The thermal steam reforming option was not considered further since in-house experimental evidence indicates a high catalyst temperature, in excess of 1800°F, is required to achieve high fuel conversion and carbon-free operation. The level of catalyst temperature to achieve the heat transfer characteristics necessary for high conversion implies very high tube wall temperatures with associated material structural problems.

Conventional Adiabatic Reformer

A functional schematic of the conventional adiabatic reformer power plant is shown as Figure 3-2.

Description of Fuel Processing Subsystem - In the conventional adiabatic reformer, preheated steam and air are combined with the fuel providing, by combustion, the endothermic heat of reforming in the catalyst bed. Previous experimental work has indicated that reactor exit temperatures in the 1650°F range are required to achieve high fuel conversion. These high temperatures are necessary to compensate for deactivation of the catalyst by the sulfur in the feed. To achieve high exit temperature, it is necessary to either preheat the steam and air to high temperatures or to use high air-to-fuel ratios. The latter option involves additional combustion and therefore, lower hydrogen production and efficiency. For this study, the steam was preheated to 1450°F, the air to 1227°F, and air was added in the amount of 0.425 moles oxygen (in air) per atom of carbon (in fuel).

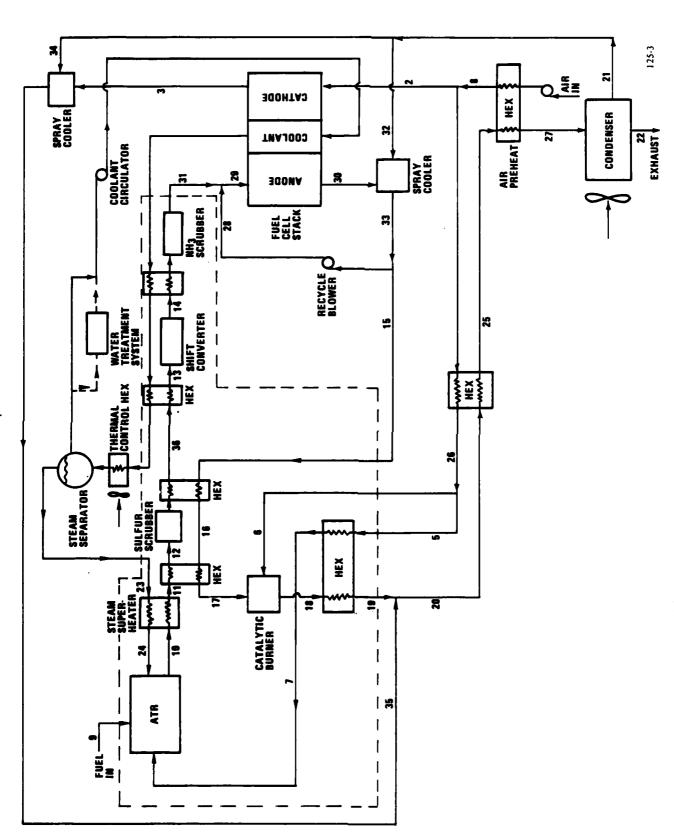


Figure 3-2. Conventional Adiabatic Reformer

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This adiabatic operating point assures carbon-free operation and eliminates the need for any unsafe fuel-to-air heat exchangers.

In Figure 3-2, the area enclosed by the dashed lines represents the fuel processing subsystem. A functional description of the subsystem follows:

The adiabatic reformer product fuel gas at station 10 is cooled from 1650°F to 488°F to effect sulfur removal via the following reaction: ZnO + H_2S → ZnS + H_2O . The zinc oxide (ZnO) cannot be regenerated with present technology. Due to the high amounts of sulfur contained in logistic fuels, it is anticipated that a large amount of ZnO will be required. After leaving the reformer, heat is transferred from the hot product gas to first superheat steam for the reformer and secondly to preheat anode exhaust gases prior to catalytic combustion. Desulfurized gases exiting the zinc oxide beds are further cooled to 391°F for entrance to a lowtemperature shift conversion reactor at station 13. In this reaction, carbon monoxide is converted to hydrogen via the water gas shift reaction: CO + H₂O → H₂ + CO2. This increases the availability of hydrogen for electrochemical conversion in the fuel cell, increasing both power plant efficiency and fuel cell performance. Shifted gases at station 14 are cooled to 400°F to effect removal of any ammonia formed in the reformer. In a separate scrubber upstream of the cell, ammoniacontaining fuel gases are passed over phosphoric acid (in charcoal support) to form di-ammonium phosphate. The scrubbers are replaced periodically. If ammonia removal was not provided for, the same reaction would occur inside the cell resulting in performance decay with time. The fuel gases at station 31 are now completely processed for entrance to the fuel cell section of the power plant.

Description of Balance of Power Plant - Due to the low hydrogen content (34% by vol) of the processed fuel gas, it is desirable to utilize a very high percentage of the hydrogen in these gases in order to maximize power plant efficiency. An overall utilization of 94% of the hydrogen in these gases was assumed for this study. Since the actual utilization of hydrogen across the anode of the fuel cell, itself, is limited by gas distribution considerations to the 80-85% range, this overall utilization is effected by means of an anode recycle, wherein anode exhaust gas

is recycled back to the anode inlet (station 28). If anode recycle is not utilized, the overall power plant efficiency would be reduced from 33% to 28%. Lean anode exhaust gases are preheated to $1044^{\circ}F$ prior to mixture with air (station 6) and catalytic combustion. Combustion must be catalytic due to the extemely lean nature $(2.3\%~H_2)$ of this fuel gas stream. Burner exhaust at station 18 is cooled by preheating air for the reformer.

Air enters the system at station 1 and is preheated to 200°F at station 8. At this point, air used in the catalytic burner and adiabatic reformer is split off from the main air line to the fuel cell. The remainder of the air at station 2 is fed to the fuel cell cathode, where oxygen is utilized by the reaction:

$$^{1}_{2}$$
 O₂ + 2H⁺ + 2e⁻ \rightarrow H₂O

Hydrogen is utilized at the anode by the following reaction:

$$H_2 \rightarrow 2H^+ + 2e^-$$

Cathode exhaust gas, containing depleted air, fuel cell product water, and some phosphoric acid mist is fed to a spray cooler where it is mixed with liquid water. This lowers the bulk temperature to 250°F where the phosphoric acid condenses. (The same type spray cooler is also included in the anode side, as indicated in Figure 3-2). The gas at station 20 is the combined cathode and anode exhaust and contains all of the water which must be recovered from the reformer and spray coolers. This mixture is cooled to preheat air for the reformer and catalytic burner and sent to a condenser at station 27.

The water treatment and power conditioning subsystems are similar to the units included in the on-site power plant and are essentially the same for all three power plants in this study.

<u>Thermodynamic Table</u> - A thermodynamic table corresponding to the schematic (Figure 3-2) is included as Table 3-1.

TABLE 3-1
ADIABATIC REFORMER THERMODYNAMIC TABLE

	TEMP	HZ	HZO	CH4	CO	COS	02	NZ	ENTH
1	95.	0.0	0.0	0.0	0.0	0.0	10.1858	37.898	186652.
2	200.	0.0	0.0	0.0	0.0	0.0	8.0020	29.766	174391.
3	400.	0.0	8.0020	0.0	9.0	0.0	4.0010	29.766	-563423.
4	200.	0.0	0.0	0.0	0.0	0.0	2.1838	8.131	47628.
5	550.	0.0	0.0	0.0	0.0	0.0	1.7872	6.648	60100.
6	550.	0.0	0.0	0.0	0.0	0.0	0.3966	1.483	13389.
7	1227.	0.0	0.0	0.0	0.0	0.0	1.7872	6.648	103475.
ੌ 8	200.	0.0	0.0	0.0	0.8	0.0	10.1858	37.898	222019.
9	77.	0.0	0.0	0.0	0.0	0.0	0.0	0.0	-30488.
_ 10	1650.	7 . 0467	_ 7.2663	0.0135	1.5622	2.6296	0.0 .	. 6.640	-842233.
11	1169.	7.0467	7.2663	0.0135	1.5622	2.6296	0.0	6.640	-949691.
12	488.	7.0467	. 7.2663	0.0135	1.5622	2.6296	0.0	6.640-	-1091159.
13	391.	7.0467	7.2663	0.0135	1.5622	2.6296	0.0	6.640-	-1110264.
14	517.		5.8003	0.0135	0.0962	4.0956	0.0	6.640-	-1110264.
15	250.	0.5108	11.1312	0.0135	0.0962	4.0956	0.0	6.640-	-1719829.
16	329.	0.5108	11.1312	0.0135	0.0962	4.0956	0.0		-1705269.
17	1044.	0.5108	11.1312	0.0135	0.0962	4.0956	0.0	-	-1563801.
18		-	11.6689	0.0	. 0.0	4.2053	0.0661		-1550412.
19	1134.	0.0	11.6689	0.0	0.0	4.2053	0.0661		-1593787.
20	612.	0.0	21.7735	0.0	0.0	4.2053	4.0671		-2402572.
21	105.	0.0	17.9467	0.0	0.0	0.0	0.0	0.0	Q.
	105.		3.8268	0.0	. 0.0	4.2053	4.0671		-903383.
23	330.	0.0	10.5132	0.0	0.0	0.0	0.0		-1014267.
24	1450.	0.0	10.5132	0.0	0.0	0.0	0.0	0.0	-906809.
25	564.	0.0	21.7735	0.0	0.0	4.2053	4.0671		-2428433.
26	550.		0.0	0.0	0.0	0.0	2.1838	8.131	73489.
27	498.	0.0	21.7735	0.8	0.0	4.2053	4.0671		-2463800.
28	250.	1.5323	33.3936	0.0405	0.2887		0.0		5159488.
29	321.	10.0450	39.1939	0.0540	0.3849	16.3823	0.0		-6269752.
30	400.	2.0431	39.1939	0.0540	0.3849	16.3823	0.0		-6257117.
31	400.	6 5127	5.8003	0.0135	0.0%2	4.0956	0.0		-1133742.
32	105.	0.0	5.3309	0.0	0.0	0.0	0.0	0.0	-622068.
33	250.	2.0431		0.0540	0.3849	16.3823	0.0		-6879185.
34	105.	0.0	2.1026	0.0	0.0	0.0	0.0	0.0	-245362.
35	265.	0.0	10.1046	0.0	0.0	0.0	4.0010		-808785.
36	414.	7.0467	7.2663	0.0135	1.5622	2.6296	0.0	6.640	-1105719.

TEMPERATURES ~ DEGREES F
FLOWS ~ LBMOLES/HR
ENTHALPIES ~ CATALYSTS MFR'S DATA

Areas of Development Risk - There are three critical heat exchangers in the system. These units see high maximum gas temperatures and operate at high effectiveness. By far the most critical of these heat exchangers is the steam superheater which sees a maximum gas temperature of 1650°F and operates at 85% effectiveness. Important design considerations for construction of this unit include material selection and allowance for thermal expansion. Other critical heat exchangers include the adiabatic air preheater with a 1322°F maximum gas temperature and 88% effectiveness and an anode exhaust preheater at 1169°F and effectiveness of 86%.

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Fuel cell performance on the very lean fuel gases (including anode recycle) assumed in this study remains to be verified by experimental data. In addition, the anode recycle results in increased acid loss due to high anode flow rates and necessitates inclusion of a recirculation pump operating on reducing gases at 250°F. The anode recycle, and its impact on technical risk, can be eliminated. This would lower the system overall efficiency by five points.

An important design consideration concerning adiabatic operation is the design of a fuel injection configuration which ensures carbon-free operation. Considerable experimental work on this subject has been done and is ongoing under Army Contract DAAK70-82-C-0012, Evaluation of Adiabatic Reformer in Mixed Gas Cycle.

Finally, design of a phosphoric acid scrubber to effect ammonia removal to desired levels while operating at 400°F, has been demonstrated on a laboratory level.

The baseline approach is to use non-regenerable zinc oxide to effect sulfur removal. From an operating and maintenance viewpoint, it would be desirable, but not necessary, to develop a regenerable system.

Hybrid Reforming

A functional schematic of the hybrid power plant is shown as Figure 3-3.

Description of Fuel Processing Subsystem - The hybrid reformer approach combines the best features of the TSR and the adiabatic reformer system to obtain an improved efficiency and a reduction in development risk. In the hybrid system, fuel and steam are fed to a tubular (primary) reactor containing a nickel-free calcium aluminate catalyst which tends to inhibit carbon formation. Only partial conversion of the fuel is achieved in this reactor. The process heat required is provided by heat transferred to the tubes from the hot burner gases. A small amount of air $(O_2/C \text{ ratio} = 0.21 \text{ as opposed to } 0.435 \text{ for adiabatic reformer})$ is then added to the partially converted fuel gas and the fuel conversion is completed in the adiabatic (secondary) reactor. The remainder of the fuel processing subsystem, downstream

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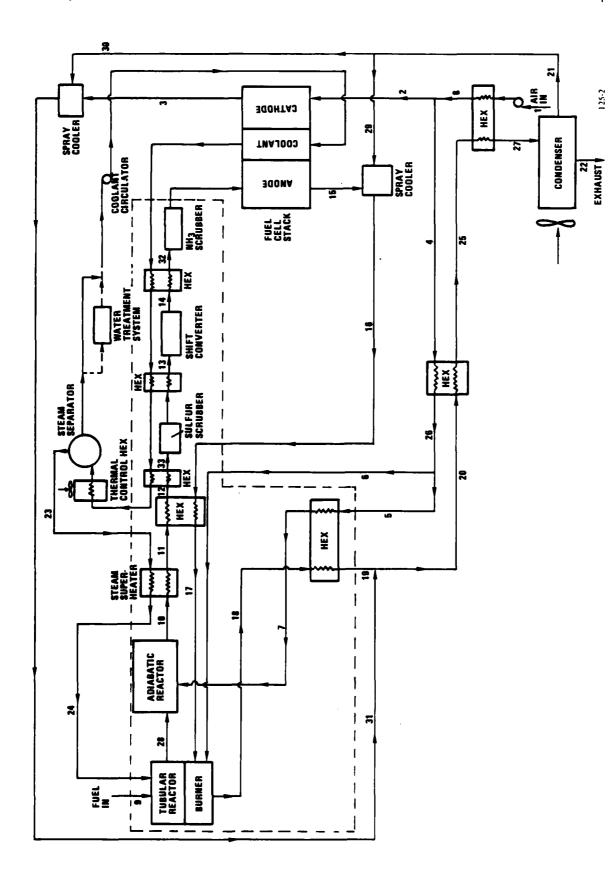


Figure 3-3. Hybrid System

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of the adiabatic reactor, is similar to the corresponding subsystem in the adiabatic reformer power plant.

<u>Description of Balance of Plant</u> - Due to the much lower air-to-fuel ratio that the hybrid processor operates at relative to the adiabatic reformer system, resultant fuel cell gases are richer in hydrogen (43% vs 34% for the adiabatic reformer system). This allows the cell stack to operate at 78% hydrogen utilization without the need for any anode recycle.

Except for certain differences in heat exchanger operating conditions (to be discussed in the Section 6.3.4) the balance of plant is very similar in function to that in the adiabatic reformer system.

<u>Thermodynamic Table</u> - A thermodynamic table corresponding to the schematic (Figure 3-3) is included as Table 3-2.

<u>Areas of Development Risk</u> - To ensure carbon free operation, temperature at the exit of the tubular reactor must be in the 1750°F range. Since heat must be transferred through a tube wall in this reactor, this implies tube wall temperatures in the 2000°F range. Selection of proper tube wall materials to meet this requirement for long-term operation is critical.

The uncertainties of the fuel cell stack in the adiabatic reformer system, caused by inclusion of the anode recycle, are eliminated for the hybrid system.

Heat exchanger requirements are similar, but somewhat less stringent, than in the adiabatic reformer case. Requirements for the steam superheater are identical, but the reformer air preheater operates at 1100°F maximum gas temperature and only 58% effectiveness and the anode exhaust preheater at 1038°F and effectiveness of 87%.

Requirements for sulfur and ammonia removal are similar to that in the adiabatic reformer power plant.

TABLE 3-2
HYBRID THERMODYNAMIC TABLE

	TEMP	HZ	HZO	CH4	co	cos	02	N2	ENTH
_ 1	95.	0.0	0.0	0.0	0.0	0.0	9.7753	36.390	179205.
2	200.	0.0	0.0	0.0	0.0	0.0	7.5402	28.048	164327.
3	400.	0.0	7.5402	0.0	0.0	0.0	3.7701	28.048	-530907.
4	200.	0.0	0.0	0.0	0.0	0.0	2.2351	8.342	48833.
5	500.	0.0	0.0	0.0	0.0	0.0	0.8330	3.099	26581.
~ 6	500.	0.0	0.0	0.0	0.0	0.0	1.4021	5.243	44919.
7	848.	0.0	0.0	0.0	0.0	0.0	0.8330		36722.
8	200.	0.0	0.0	0.0	0.0	0.0		36.390	213161.
9	77.	0.0	0.0	0.0	0.0	0.0	0.0	0.0	-28758.
10	1650.	8.1661	7.2898	0.0269	1.6034		0.0	3.095	-841186.
	1160.	8.1661	7.2898	0.0269	1.6034	2.3364	0.0	3.095	-939292.
12	549.	8.1661	7.2898	0.0269	1.6034	2.3364	0.0	3.095-	-1053617.
13	373.	8.1661	7.2898	0.0269	1.6034	2.3364	0.0	3.095-	-1084829.
14	517.	9.6669	5.7890	0.0269	0.1026	3.8372	0.0	3.095-	-1084829.
15	400.	2.1267	5.7890	0.0269	0.1026	3.8372	0.0	3.095-	-1150393.
_16	250.			0.0269	0.1026	3.8372	0.0	3.095-	-1261867.
17	1060.		6.7443	0.0269	0.1026	3.8372	0.0	3.095-	1147542.
18	1075.	0.0	8.9248	0.0	0.0	3.9667	0.2337	8.338-	-1319289.
	1024.		8.9248		0.0	3. 9 667	0.2337	8.338-	-1329431.
20	545.	_0.0	_18.6754_	_0.0	0.0	3.9667	4.0038	36.387-	-2118278.
21	105.	0.0	15.0657	0.0	0.0	0.0	0.0	0.0	0.
22	105.	0.0				3.9667			-849102.
23		0.0	11.9000	0.0	0.0	0.0	0.0	0.0 -	-1148056.
-	1250.		11.9000	.0.0 ,	0.0 :_		0.0	0.0 -	-1049949.
25	499.	0.0	18.6754	0.0	0.0	3.9667			-2140945.
26		0.0	0.0	0.0	0.0	0.0	2.2351		71500.
27	430.	0.0	18.6754	0.0	0.0	3.9667	4.0038	36.387-	-2174901.
28	1750.		. 8.4618	1.8445	0.8061	1.3160	0.0		-869975.
29	105.	0.0	0.9553	0.0	0.0	0.0	0.0	0.0	-111474.
30	105.	0.0	2.2104	0.0	0.0	0.0	0.0	0.0	-257941.
31	250.	0.0	9.7506	0.0	0.0	0.0	3.7701	28.048	-788848.
. 32	_ 400.	9.6669		0.0269	0.1026	3.8372			-1105970.
33	478.	8.1661	7.2898	0.0269	1.6034	2.3364	0.0	3.095-	-1066330.

TEMPERATURES ~ DEGREES F FLOWS ~ LBMOLES/HR ENTHALPIES ~ GIRDLER BASED

Cyclic Reforming

A functional schematic of the cyclic power plant is shown as Figure 3-4.

<u>Description of Fuel Processing Subsystem</u> - The cyclic reforming concept integrates all of the fuel processing functions necessary to process logistic fuels into a gas deliverable to a fuel cell (except low-temperature shift conversion since this catalyst is poisoned by sulfur). First, the fuel is vaporized and steam is preheated. When the fuel/steam mixture reaches approximately 1000°F, the gases pass over a calcium aluminate thermal reforming catalyst and partial fuel conversion is achieved.

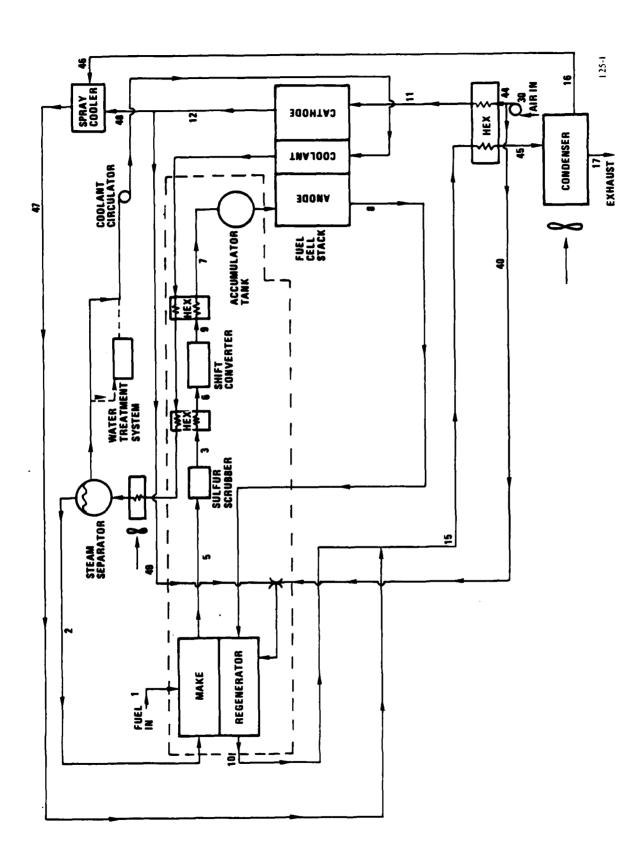


Figure 3-4. Cyclic System

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The gases, which have been heated to around 1800°F at this point, are then routed over a conventional nickel steam reforming catalyst to complete fuel conversion. The final steps are gas cooling followed by high temperature shift conversion to partially convert carbon monoxide and water to hydrogen. These functions are accomplished in two beds which are cycled periodically between a "make" mode in which fuel product gas is formed and a "regenerate" mode in which heat is provided by combustion of anode exhaust to maintain bed temperature sufficiently high to achieve proper conversion.

Figure 3-5 and 3-6 are diagrams of a typical cyclic bed configuration. As shown in Figure 3-5, steam is partially superheated in the initial ceramic packing section during the make cycle. Fuel is then injected and vaporized, slightly cooling the resultant mixture which is further superheated in the remainder of the section. The "make" gas then passes over a calcium-based catalyst section (similar to the catalyst used in the tubular reactor of the hybrid system) where partial conversion of the fuel takes place. After passing over some additional ceramic packing (over which some combustion occurs during the regeneration mode), the gas enters a catalytic steam reforming section where additional conversion occurs in the presence of a nickel catalyst. The gas passes over another burn zone, where fuel rich combustion occurs during regeneration. The gas then enters a final equilibration zone of catalytic steam reforming catalyst where final conversion takes place. The gas is then cooled in another ceramic packing zone and passed over high-temperature shift catalyst where the final "make" gas product is produced.

As shown in Figure 3-6, flow is switched in the beds, and anode exhaust gases are introduced countercurrent to the direction of the "make" gas flow during the regeneration cycle. Fresh air is introduced at this point.

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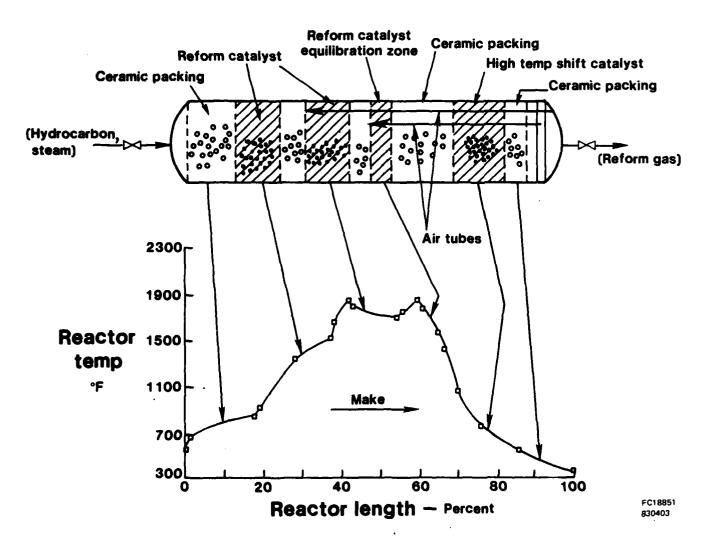


Figure 3-5. Cyclic Reformer "Make" Mode

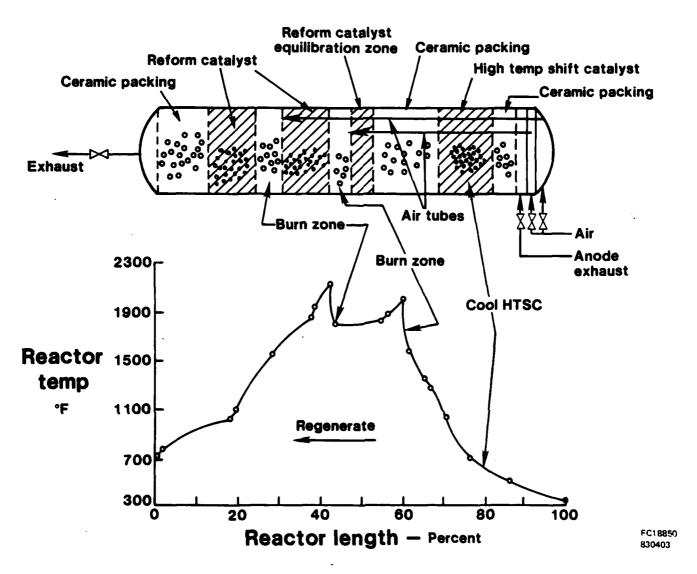


Figure 3-6. Cyclic Reformer "Regenerative" Mode

These oxidant gases are contained within tubes to prevent oxidation of the catalyst layers. The gases are then preheated, undergo staged combustion, (fuel rich over the catalytic steam reforming section) and are cooled to burner exhaust temperatures. It is estimated that approximately half of the sulfur in the fuel will be exhausted from the reactor during regeneration, as sulfur dioxide. The catalyst and ceramic beds' temperatures swing during the "make - regeneration" cycle. This is indicated by the darker area in Figure 3-7.

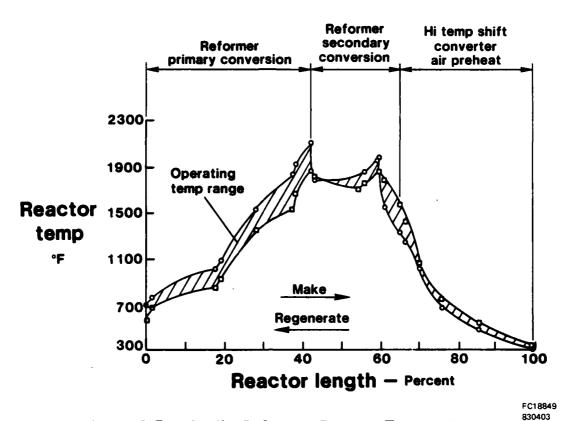


Figure 3-7. Cyclic Reformer Reactor Temperature

The remainder of the fuel processing system consists of zinc oxide beds, approximately of one half capacity to that in the adiabatic and hybrid system, two small heat exchangers, a low temperature shift converter downstream of the zinc oxide bed, and a gas storage tank to provide fuel gas during bed switchover. Unlike the other two systems, there are no high temperature or high effectiveness heat exchangers required elsewhere in the overall power plant system.

Description of Balance of Plant

The fuel cell, water recovery and treatment and power conditioning subsystems are similar to those included for the hybrid power plant.

Thermodynamic Table

A thermodynamic table corresponding to the schematic (Figure 3-4) is included as Table 3-3.

TABLE 3-3
CYCLIC THERMODYNAMIC TABLE

	TEMP	HZ	H2O	CH4	CO	CO2	02	N2	ENTH
1	77.		0.0					0.0	
2		0.0							-1202599.
3	478.	9.5599	6.0559	0.0003	0.5152	2.9471	0.0	0.0	-1011963.
4	655.	8.0	12.4654	0.0	0.0	0.0	0.0	0.0	-1168196.
. 5	478.	9.5599	6.0559	0.0003	0.5152	2.9471	0.0	0.0	-1011963.
6	471.	9.5599	6.0559	0.0003	0.5152	2.9471	0.0	0.0	-1013078.
7	400.	9.9796	5.6361	0.0003	0.0954	3.3669	0.0	0.0	-1031102.
. 8	400.	2.5086	5.6361	0.0003	0.0954	3.3669	0.0	0.0	-1075118.
9	517.	9.9796	5.6361	0.0003	0.0954	3.3669	0.0	0.0	-1013078.
10	650.	0.0	9.7246	0.0	0.0	3.4626	0.1303	8.357	-1397122.
11	200.	0.0	0.0	0.0	0.0	0.0	7.4711	28.091	163060.
12	400.	0.0	7.4711	0.0	0.0	0.0	3.7355	28.091	-525728.
13	0.	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.
14	0.	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.
15	422.	0.0	17.3374	0.0	0.0	3.4626	3.0761	30.509	-2012727.
16	107.	0.0	14.1864	0.0	0.0	0.0	0.0	0.0	8.
17	107.	0.0	3.1510	0.0	0.0	3.4626	3.0761	30.509	-748376.
40	95.	0.0	0.0	0.0	0.0	0.0	0.6431	2.418	11800.
41	0.	0.0	0.0	0.0	0.0	6.0	0.0	0.0	8.
42	0.	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.
43	0.	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.
44	95.	0.0	0.0	0.0	0.0	0.0	7.4711	28.091	137084.
45	360.	0.0	17.3374	0.0	0.0	3.4626	3.0761	30.509	-2038704.
46	114.	0.0	1.7211	0.0	0.0	0.0	0.0	0.0	-201017.
47	250.	0.0	7.6128	0.0	0.0	0.0	2.9459	22.153	-615605.
46	400.	0.0	5.8917	0.0	0.0	0.0	2.9459	22.153	-414588.
49	400.	0.0	1.5794	0.0	0.0	0.0	0.7897	5.939	-111139.

TEMPERATURES ~ DEGREES F FLOWS ~ LBMOLES/HR ENTHALPIES ~ GIRDLER BASED

Areas of Development Risk

The cyclic reformer, while indicating the most potential for high efficiency and system simplicity outside of the fuel processor itself, is the least developed of the fuel processors considered in this study, and therefore the level of technical risk represents an unknown. The risks include the following: long-term operability under thermal cycling operation in the presence of sulfur, control valve complexity and reliability, prevention of net carbon build-up, and adequate control of reactant fuel gas pressure during transient operation.

Concerns outside of the fuel processor subsystem are minimized relative to the other two power plants. Fuel cell stack concerns found in the adiabatic reformer system are eliminated. Heat exchangers are not a critical design issue for this power plant. Finally, sulfur removal requirements are reduced and ammonia removal is not required.

COMPARISON OF REFORMER OPTIONS

The characteristics of power plants based on each of the alternative fuel processor concepts were estimated. The characteristics of the power plant were then compared. A comparison was made of power plant efficiency, system complexity, and manufacturing cost. A 100-kW power plant incorporating a conventional steam reforming fuel processor served as the basis for the estimating characteristics of power plants incorporating the alternative fuel process types. All power plant components with the exception of the fuel processing units and related thermal control heat exchangers, controls, and pallet structure/piping were assumed the same for each power plant. The necessary fuel processor modifications for the handling of diesel fuel were then defined.

Each of the three comparisons is discussed below.

Efficiency Comparison

The power plant comparisons show the cyclic reformer to have the greatest potential for reforming diesel fuel at high system efficiencies. See Table 3.4 below. A fuel cell power plant system based on the cyclic reformer has the potential for an overall electrical efficiency of 40%.

TABLE 3.4. EFFICIENCY COMPARISONS

Parameter	ATR (Anode Recycle)	ATR (No Anode Recycle)	Hybrid	Cyclic
Efficiency	33%	28%	35%	40%
Relative Complexity	1.0	1.0	1.0	1.25
Relative Manufacturing Cost	1.0	1.0	1.0	1.0

This efficiency is at least five percentage points better than the hybrid system, and seven paints higher than the autothermal reformer. This high efficiency is clearly an advantage for reducing a life cycle cost of providing power at remote sites.

Reliability Comparison

The three candidate fuel processing subsystems were compared from the viewpoint of their potential reliability. The criteria used to quantify their reliability were mean-time-between-failures (MTBF) and failure rate (failure/ 10^6 hours).

The three fuel processing subsystems are in a conceptual phase, and therefore are only defined by their major components, e.g., catalyst beds and heat exchangers. Experience has shown the static devices to be very reliable; fuel processing sub-

3

system failures are primarily associated with the dynamic components. Therefore, to make the reliability comparison meaningful, the necessary valves and sensors required to make each subsystem function were assumed.

With each subsystem thus defined, the failure rate of each component was estimated using electric utility experience as published in the following documents:

- A. IEEE Guide to the Collection and Presentation of Electrical, Electronic, and Sensing Component Reliability Data for Nuclear-Power Generating Stations, IEEE STD 500-1977.
- B. Nuclear Plant Reliability Data System 1979 Annual Reports of Cumulative System and Component Reliability, NUREG/CR-1635, Southwest Research Institute.

By summing the failure rates of all components in a subsystem, the subsystems were thus compared. This analysis indicated that failure rates for the adiabatic and hybrid fuel processing subsystems were comparable, while the failure rate for the cyclic subsystem was twice that of the hybrid and adiabatic fuel processing subsystems. This translates into a MTBF for the cyclic subsystem at one-half that of the other two subsystems.

Previous studies have shown that a fuel processing subsystem contributes approximately 20 percent to a typical fuel cell power plant's failure rate. A power plant, therefore, with a cyclic fuel processing subsystem would have a failure rate (λ) and mean-time-between-failure of 1.20 and 0.80 respectively compared with power plants with adiabatic and hybrid subsystems. The comparsion is shown in Table 3-5.

The lower reliability of the cyclic fuel processing subsystem is due to the additional valving and switching required to manipulate the complex make/regenerate reforming process. If early technology base programs confirm means to handle the process complexity then other attributes of the cyclic subsystem make it desirable for consideration.

TABLE 3-5. FUEL PROCESSOR SUBSYSTEM RELIABILITY ATTRIBUTE COMPARISON

	Relati	∨e	
	λ	MTBF	
Fuel Processing Subsystem Basis			
Adiabatic	1.0	1.0	
Hybrid	1.04	0.96	
Cyclic	2.03	0.49	
Power Plant Basis			
Adiabatic	1.0	1.0	
Hybrid	1.01	0.99	
Cyclic	1.24	0.81	

Cost Comparison

The comparison of the relative manufacturing cost of the power plants based on the alternative fuel processors is listed below. These were compared with a conventional steam reforming power plant system.

TABLE 3-6. COST COMPARISON

Power Plant Description	Relative Cost
Conventional Steam Reformer*	1.00
Adiabatic Unit	1.06
Cyclic Unit	0.99
Hybrid Unit	1.07

*Baseline

The manufacturing cost of a fuel processor subsystem that includes a cyclic reformer is 1 percent lower in costs than the baseline gas utility fuel cell power

plant and has a comparable overall system efficiency. The cyclic reformer portion of this subsystem was estimated to be roughly twice the baseline power plant manufacturing cost. However, the balance of major fuel processor components needed to complete a fuel processor subsystem are fewer in number when compared to the other reformer approaches. Process valving required for cyclic reformer control adds little to the overall power plant cost.

Manufacturing costs for fuel processor subsystems containing the adiabatic and hybrid reformers are estimated as 7 and 8 percent respectively above the cyclic concept. The higher cost is attributed to process components that have to be added to the fuel processor and power section subsystems to accommodate the diluting and contaminating effects of continuously burning fuel with air in the reformer process stream. Other components have been added to the subsystem to increase fuel, air, and steam temperatures before being supplied to the reformer.

Selection of Preferred Option

Discussion of the relative importance of power plant characteristics with remote site power plant users indicates that the cost of full supply and overhaul and maintenance costs are all important in prime mover selection. As a result the cyclic reformers high efficiency makes it a preferred choice. Although its relatively low MTBF augurs against its selection, MTBF can be improved by development and redundancy reducing the tendency to high overhaul and maintenance. Assessment of the reliability of the cyclic reformer's reliability potential requires additional evaluation, both analytical and experimental.

Therefore because its efficiency potential is significantly higher than the alternative fuel processing concepts and its reliability has the potential to be improved, the cyclic reformer has been selected for exploratory development.

Development Plan

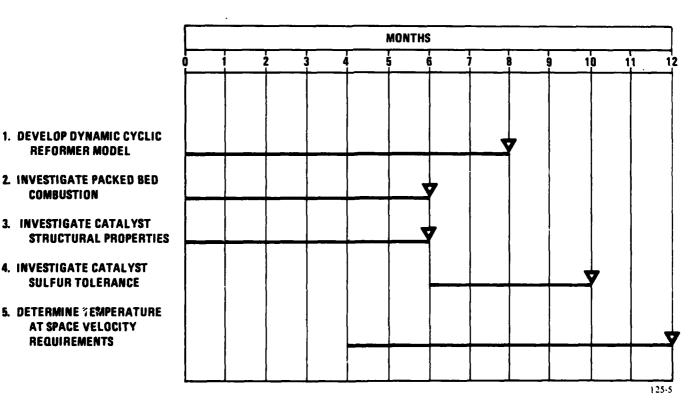
REFORMER MODEL

SULFUR TOLERANCE

AT SPACE VELOCITY REQUIREMENTS

COMBUSTION

Development of a reformer capable of processing diesel fuel would be an exploratory type of program, and the results would enable a reformer concept selection verification. To achieve that object, a series of exploratory test programs have been defined to evaluate underlying technology assumptions used as the basis for the cyclic reformer system concept analytical comparisons. The overall program shown in Figure 3-8, planned for 12 months, allows 50 percent completion of Task 1.0 before initiating certain tests. The resulting analytical data will provide a basis for planning and implementing Task 4 and 5 testing. The selected exploratory programs, shown in Table 3-7, are listed in order of importance to the reformer concept selection process. Completion of the recommended exploratory program would provide the necessary understanding to proceed with a fuel processor subsystem brassboard design and subscale test program.



- Figure 3-8. Technology Evaluation Program
 - 3-23

TABLE 3-7. TECHNOLOGY EVALUATION PROGRAM

Task No.	k Task	Reason	Benefit	B&P Cost
-	Modify existing power plant system analytical models to consider the dynamic cyclic reformer subsystem operated on logistic fuels.	Identify system components and control logic required to satisfy fuel cell power plant operating require- ments.	Verify feasibility of cyclic reformer operation with logistic fuels in a fuel cell power plant system. Provide critical data for technology testing.	195 K
~	Investigate combustion in a regenerable cyclic reformer packed bed.	Need to achieve a uniform catalyst bed temperature profile.	Achieve minimum catalyst temperature consistent with high power plant efficiency.	185 K
က်	Determine cyclic reformer catalyst and insulation system structural properties when subjected to thermal and process composition cycling.	Verify structural stability of commercially available reformer catalyst and insulation when subjected to cyclic exposure.	Provide basis for selecting cyclic reformer materials.	200 K
4.	Investigate temperature and sulfur tolerance of final conversion catalyst (reformer exit).	Determine operating tem- perature and space velocity requirements for catalyst.	Achieve high conversion at the optimum reformer efficiency for the power plant system.	100 K
လ	Investigate primary gasification catalyst carbon formation tolerance, regeneration characteristics and determine operating temperature/space velocity requirements.	Determine minimum bed size to achieve desired heavy fuel conversion.	Achieve required conversion to obtain maximum reformer efficiency and stable operation.	150 K
			Total	\$830 K

PROGRAM APPROACH

The technology evaluation program, shown in Table 3-7, has been configured to investigate a variety of reformer component questions. The necessary data will be obtained to confirm the cyclic reformer selection for future developments needed to satisfy Air Force requirements for higher efficiency and good power plant utility. The anticipated approach for implementing the selected program is discussed in the following sections.

The first step for any of the five tasks involves defining the specific technical issues to be investigated, establishing appropriate test rig configurations, and determining operating ranges and measurements required to analyze each technical risk issue. This includes the number of cycles, test hours, temperatures, process gas analysis and/or stability tests.

The Task 1, analytical model effort will be initiated at the start of the twelvemonth program. This effort will include the conversion of existing steady state reformer process analytical models to represent a thermally transient process, which is characterized of the cyclic reformer. A new analytical model will be created which will describe the transient heat and mass transfer characteristics of the various packing and catalyst materials in the cyclic reformer. This analysis will include a kinetic description of heavy fuel cracking, to light products and subsequent reforming to H₂, CO, CO₂. The analytical technique required for proper description of the cyclic reformer have been used extensively in existing PSD fuel processing and system analysis models. Results from the Task 1 effort would be utilized to define the specific approach to Task 2 through 5.

The Task 2 investigation of temperature profiles in a packed bed resulting from combustion would involve 10 hours of testing for each of the selected mechanical configurations. A comparison would be made of temperature measurements in combination with burner heat flux and the results would demonstrate temperature profiles resulting from direct firing of a packed bed.

*

<u>...</u>

The Task 3 catalyst and insulation structural properties investigation is mechanical in nature and can be implemented independent of Task 1. The investigation would involve a system of materials including an internally insulated vessel (simulated), state-of-the-art ceramic insulation, selected catalysts, and ceramic packing. The rig would be instrumented for temperature cycling through temperature swings of 250°F. The anticipated period for each cycle is two minutes. Half of the fuel processor cycle involves regeneration and heating while the other half involves fuel conversion. Each catalyst/insulation configuration would be periodically inspected to document physical integrity of insulation, catalyst, and the overall dimensional stability of the containment vessels and internal supports. After test inspections would evaluate the condition of test materials.

Tasks 4 and 5 are performance tests monitored by thermal instrumentation and gas sampling. These tasks will focus on process performance characteristics for candidate catalysts and packed bed materials. In general, 50-hour screening tests are anticipated to identify leading candidate catalyst; this will be followed by a final 1,000-hour performance stability test for the leading candidate. A test rig containing final conversion catalyst is required for Task 4 and another containing candidate primary gasification catalyst is envisioned for Task 5.

Test rigs for Tasks 2 thru 5 will be specifically designed to explore the specific area associated with each task listed in Table 1. Appropriate rig scale selections will be made considering heat loss and its effect on the process flow dynamics and mechanical representation of ultimate subsystem components. Once the rig. has been defined for each technology evaluation program task, the measurements will be determined to complement the Task 1 analytical modelling and instrumentation selected. Finally, a sequential test method will be written to guide each exploratory test.

The combined results of testing accomplished with Tasks 2 thru 5 and the completion of Task 1, Development of Transient Reformer Process Model, will provide a basis for comparing the results from the test programs. The analytical models would be used in conjunction with the technical data generated from the test pro-

grams to confirm the cyclic reformer selection and identify other technical areas requiring exploratory investigation with a brassboard rig simulation of the fuel processor subsystem.

ATTACHMENT I

DD Form 882

	(Pursu	REPORT OF INVENTIONS (Pursuant to "Patent Rights" Contract CI	F INVENTIONS AN	AND SUBCONTRACTS ause) (See Instructions on Reverse Side)	(e)		FORM A	FORM APPROVED OMB NO. 22-R160	
1. NAME AND ADDRE	SS OF CONTRAC	NAME AND ADDRESS OF CONTRACTOR (Include 219 Code)				2. CONTRACT NUMBER	e e		
Power	Power Systems Division	ivision	Ę			DAAK 70	70-82-C-0012		
P.0.	Box 109					3. TYPE OF RE	TYPE OF REPORT (Check One)		
South	Windsor,	South Windsor, Connecticut O	. +12090			- INTERIM	¥	FINAL	
			SECTION	SECTION 1 - INVENTIONS ("Subject Inventions")					
A. INVENTION DATA (Listed below are	INVENTION DATA (Listed below are all inventions required to be reported) (II "Wone," so state)	o be reported) (11 "None,	** 50 state)			1		
(I) NAME OF INVENTORISI	(#) TORIS		TITLE OF INVE	INVENTION	(111) CONT DISC IDENTIFIC OR PATENT	III) CONTRACTOR DISCLOSURE IDENTIFICATION NUMBER OR PATENT APPLICATION SERIAL NUMBER	(19)CONTRACTOR ELECTS TO FILE U. S. PATENT APPLICATION	(v)CONFIRM LICENS LICENS FORWARDIN CONTRAC OFFIC	Z Z Z Z Z Z Z Z Z Z Z Z Z Z Z Z Z Z Z
None									
			SECTION II - SUBCON	CONTRACTS (Containing a "Patent Rights" Clause)	Hause)				
5. SUBCONTRACT DATA (Listed is information	TA (Listed is in		I previously reported for	but not previously reported for Subcontracts) (11 "Wone," so state)					
(1) NAME AND ADDRESS OF SUBCONTRACTOR (Include Zip Code)	DDRESS RACTOR Code)	SUBCONTRACT NUMBER	(III) SUBCONTRACT PATENT RIGHTS CLAUSE	(IV) WORK TO BE PERFORMED UNDER SUBCONTRACT	INDER SUBCONTR		(V) SUBCONTRACT	COMPLETION	NOI
None									
CONTRACTOR CERT	PIET THAT PRO	NOTA DISTINGUE TOWN	SEC	SECTION III - CERTIFICATION CONTRACTOR CERTIFICS THAT PROMPT IDENTIFICATION AND TIMELY DISCLOSUBLECT INVENTIONS DEPOCED LAND BREADED INVENTIONS DEPOCED LAND TIMELY DISCLOSUBLECT INVENTIONS DEPOCED LAND TIMELY DISCLOSUBLE DISCLOSUBL					
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APPENDIX A

OPERATING CHARACTERISTICS OF THE ADIABATIC REFORMER

The operation of the adiabatic reformer has been described in previous reports (1, 2). To aid in evaluating the test results with military logistic fuel, the important features of the development of the reformer to process No. 2 fuel oil for commercial application have been summarized below.

A schematic of the bench scale test reactor is shown in Figure A-1. Prevaporized fuel and steam are rapidly mixed with air and additional steam in the mixing nozzle. The well-mixed process steam enters the catalyst bed where a complex sequence of combustion and reforming processes occur in the inlet catalyst. Carbon may accumulate in this section. Finally, in the exit section steam reforming of the residual fuel is completed.

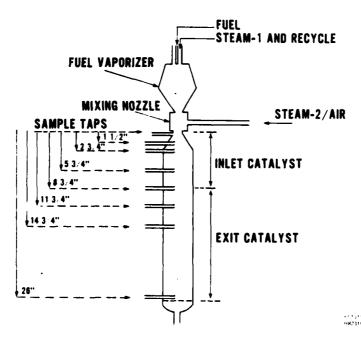


Figure A-1. Schematic of Bench Scale Adiabatic Reformer

Analytical studies of the adiabatic reformer for the 4.8-MW phosphoric acid fuel cell power plant were used to define operating conditions which would minimize system cost at a design heat rate of 9330 Btu/kWh. The operating conditions

which were set as goals for reactor development, shown in Table A-1, were selected to give minimum values for air addition (O_2/C ratio) and pre-reaction temperature while maintaining high fuel conversion.

TABLE A-1. ADIABATIC REFORMER PERFORMANCE

		Reactor	Performance
	Design(1) Baseline	Commercial(2) Nickel Catalyst	Metal Oxide(3) Plus Advanced Nickel
O ₂ /C Mole Ratio	0.36	0.42	0.36
Pre-Reaction Temperature, °F	1360	1360	1360
Exit Temperature, °F	1700	1750	1700
Conversion	98.2	99.0	>98.2(4)
Space Velocity, lbs fuel/ft ³ reactor-hr	12	24	12

⁽¹⁾ For 4.8-MW Phosphoric Acid Power Plant at 9300 Btu/kWh.

Early reactor configurations with commercial nickel catalyst required air in excess of the design value to prevent carbon laydown in the reactor entrance. This excess air reduces the quantity of hydrogen produced and thus reduces power plant efficiency. It was recognized that rapid and efficient mixing of the air and fuel was important to limit the extent of carbon-forming reactions. Therefore, a study of the effect of reactant nozzle configuration on carbon formation was made. The 2-inch diameter bench-scale reactor processing two pounds of fuel per hour was used. The nozzle configuration was varied while the fuel (No. 2 fuel oil) and the catalyst (a commercial nickel catalyst) were not changed. After testing many configurations, an optimized nozzle geometry was defined which was subsequently

^{(2) 6} inch diameter reactor with optimized nozzle.

^{(3) 2} inch diameter reactor with optimized nozzle.

⁽⁴⁾ Extrapolated to design space velocity.

scaled-up for a 6-inch diameter reactor flowing 10 pounds of fuel per hour. The ability to scale up the nozzle design was demonstrated by a close agreement in the minimum O_2/C requirement for the two reactors. However, the reactor with optimized nozzle was still limited in performance by carbon formation. As shown in Table A-1, the minimum O_2/C requirement exceeded the design goal.

The effect on carbon formation of variation in catalyst formulation was therefore studied while holding the nozzle configuration fixed. From the position of the carbon deposited in the reactor and from the variation of product composition with reactor length, it was apparent that the principal function required of the catalyst depended on its position in the reactor bed; in the inlet section where combustion reactions predominated, the ability to inhibit carbon accumulation was paramount; in the exit section, the ability to reform residual methane was the only requirement. Hence, for carbon formation studies, the catalyst in the inlet of the 2-inch diameter reactor was changed while leaving the exit catalyst in place. In this way the effect of changes in the inlet catalyst could be rapidly determined.

Experiments in a laboratory microreactor had suggested that metal oxide catalysts would show superior resistance to carbon formation. When these catalysts were placed in the reactor it was clear that a major change in the rates of reaction in the combustion zone had occurred, as evidenced by a change in the temperature and product composition profiles from those observed with the commercial nickel catalyst. At the same time the minimum value for O_2/C at which the reactor would operate carbon-free was reduced.

In Figure A-2, the characteristic carbon-free operating regime of the reactor is illustrated. At fixed pre-reaction temperature, it was found that the O_2/C ratio in the feed could be lowered to a point where increasing pressure drop across the reactor indicated carbon formation; raising the O_2/C ratio from this value reversed the pressure increase. Using this technique at different pre-reaction temperatures, a reactor operating line, above which the reactor could operate carbon-free, was defined. For reactors filled with commercial nickel catalyst, this line has a characteristic slope. Improvement in reactor performance was indicated by a lower value for the O_2/C intercept of the operating line. Metal oxide catalyst can be

see in Figure A-2 to have extended the carbon-free operation of the reactor below that obtained with commercial nickel catalyst. The slope of the operating line for the metal oxide catalyst was similar to that of the nickel catalyst. More importantly, metal oxide permitted operation of the reactor at the baseline design value for O_2/C .

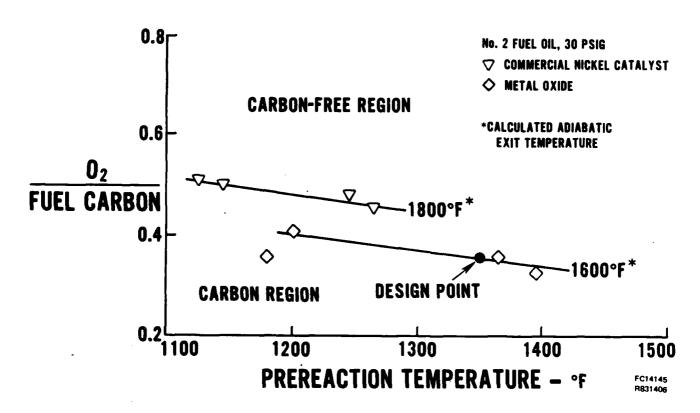


Figure A-2. Effect of Catalyst on Carbon Formation in the Adiabatic Reformer

The fuel cell power plant not only requires that the reformer operate carbon-free, but also that it achieve high fuel conversion at the design conditions. For a given catalyst in the exit section of the reactor, the conversion had been shown to correlate with the exit temperature and was independent of whether that temperature was achieved by the addition of air (increased O_2/C) or by an increase in preheat temperature (2). With commercial nickel catalyst in the exit to the reactor, the required conversion could only be achieved at high temperatures. A more

active nickel catalyst was developed in a parallel laboratory program which when placed in the exit of the 2-inch diameter reactor, gave the conversions obtained in an earlier program (2), and shown in Figure A-3.

Since the adiabatic reformer does not require an external burner cavity, the catalyst volume can be commensurately higher than the catalyst volume in tubular, externally fired reformers for an equivalent overall volume. The design goal for the adiabatic reformer in Table A-1 was set as 12 pph/ft³. The 2-inch diameter test reactor did not contain sufficient catalyst to give space velocities less than about 30 pph/ft³. The conversion at 12 pph/ft³ was, therefore, projected from the data at 40 pph/ft³ by use of a simple first order model for methane conversion in the reactor exit. A good fit to the data was obtained and the curve calculated for the design space velocity and plotted in Figure A-3 showed that the conversion achieved by the advanced nickel catalyst projected to the design point.

A 2-inch bench-scale reactor with the optimized nozzle configuration, metal oxide catalyst in the inlet section, and the high activity nickel catalyst in the exit section was seen for 450 hours on No. 2 fuel oil in an earlier program funded by the Electric Power Research Institute. With a similiar reactor catalyst loading, a series of runs was made under Army funding. Over 1400 hours of testing were completed with Army logistic fuels at conventional adiabatic reformer conditions under that contract (2), and with the same catalyst, almost 800 hours were completed with diesel and methanol fuels at mixed gas cycle conditions under the present contract.

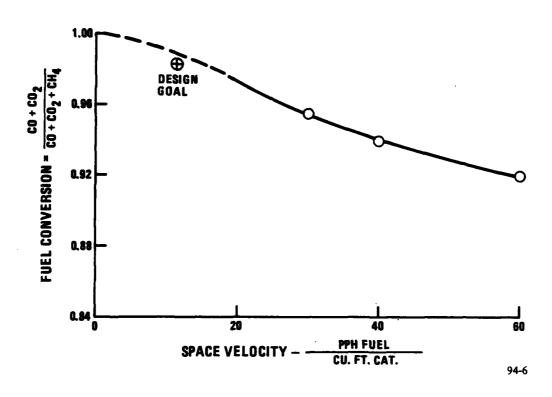


Figure A-3. Fuel Conversion as Function of Space Velocity at 1700°F Exit Temperature

MID 0.53 lbs EXIT 1.81 lbs

CATALYST LOADING:

θ' = C0+C0₂ C0+C0₂+ΣnC_n

SUMMARY OF RUN 21 DATA POINTS

ı		1		API	PEI	NDI	хв									
	Comments (0')	Initial point carbon balance only 92% fuel flow off. (0.916)	Overcorrected fuel flow, carbon forming. (0.879)	Repeat of Pt 49 Run 21. Carbon-free conversion slightly less. (0.903)		Carbon was slowly forming. (0.942)	Carbon-free, air and steam were in- creased. (0.951)	Carbon formed.	Rig shut down due to burned heater wire	Repeat of Point 4B. (0.951)	Raised preheat temperature carbon	boundary nign by 0.017 O ₂ /C. (0.95) Modified exit to run low P. Carbon free, carbon balance 948. (0.951)	Carbon formed.	Carbon-free. Carbon balance only 90%. (0.942)	Lowered H ₂ O/C, carbon formed. (0.942)	Raised air, C-free. (0.955)
,	5 .	0.85	0.866	0.882			0.910	,		0.927	926.0	0.893		0.85	0.85	0.882
Balance	(Based on Exit Gas Comp.)	0.92	96.0	0.98		0.95	96.0	•		86.0	0.97	0.94	•	0.90	0.90	0.92
Reactor	sure (PSIG)	30	30	30		30	30	30	30	93	30	12	12	12	12	12
	_	1.85	2.16	1.96		2.0	2.0	2.0	2.0	2.0	2.0	2.0	5.0	2.0	2.0	2.0
Fuel	2 2 2	Diesel	Diesel	Diesel		Diesel	Diesel	Dieset	Diesel	Diesel	Diesel	Diesel	Diesel	Diesel	Diesel	Diesel
H ₂ 0/C		4.92	3.89	4.17		1.58	1.91	1.91	1.91	1.91	1.91	1.91	1.91	1.91	1.50	1.50
0 ₂ /c		0.454	0.389	0.429		0.45	0.475	0.45	0.47	0.47	0.447	0.47	0.44	0.45	0.45	0.46
	Actual	1698	1604	1670	91	1676	1712	1691	1710	1707	1724	1707	1656	1671	1670	1699
E)A	Cal- culated	1670	1510	1610	ycle Mod	1618	1660	1605	1655	1660	1675	1635	1565	1590	1590	1623
Temperatures	Adiabatic Flame	2224	2168	2202	Mixed-Gas-Cycle Mode	2300	2286	2240	2281	2288	2325	5229	2204	2225	228C	2309
	Pre- reaction	1148	1118	1135	Ē	1197	1190	1190	1194	1201	1275	1172	1173	1175	1164	1174
·	.s	8.0	30	98		4	88	29	11	80	8	94	96.5	115	116.5	118
	Point Time (Hou	₹	18	20		2	m	₹	48	5	S	9	1	∞	94	88

(0.929)

0.847

0.91+

Diesel

1.50

0.41

526.5

SUMMARY OF RUN 21 DATA POINTS

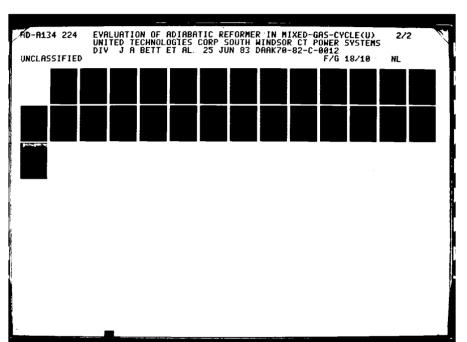
Lowered flow used W/T meter to measure exit gas flow and to calculate carbon balance. (0.951)	0.862	0.91+	12	7.5	Diesel	1.50	0.45	1696	1693	2399
Raised temperature to look at conversion. (0.948)	0.839	0.89	12	5.0	Diesel	1.50	0.45	1727	1700	2411
Carbon-free. (0.937)	0.812	0.87	Ę	5.0	Diesel	1.50	0.41	1657	1590	2321
Carbon-free. Restart of rig after one week S/D. (0.952)	0 839	0.88	=	2.0	Diesel	1.50	0.45	1719	1677	2383
Carbon-free. C-balance = 93%. (0.940)	0.874	0.93	9	5.0	Diesel	1.50	0.43	1682	1620	2338
Carbon formed. Raised T prereaction.			92	2.0	Diesel	1.50	0.45	1662	1590	2309
Turned off CO_2 . α and C-balance down by ~ 0.035 . (0.954)	0.858	06.0	15	2.0	Diesel	1.50	0.49	1741	1730	2416
increased O_2/C (catalyst temperature) and C-balance increased to 0.934. (0.958)	0.895	0.93	12	2.0	Diesel	1.50	0.49	1729	1692	2362
C-free. Repeat of Pt. 12.			12	5.0	Diesel	1.50	0.47	1705	1643	2321
Carbon formed.			12	2.0	Dieset	1.50	0.46	1683	1615	2300
Fuel F/M recalibrated, it was 48 low. (0.956)	0.901	0.90	12	2.0	Diesel	3.50	0.47	1683	1638	2318
C-formed. No gas analysis.			12	2.0	Diesel	1.50	0.40	1663	1554	2288
C-free, C-balance only 85%. Suspect gas analysis and fuel F/M. (0.936)	0.796	0.85	12	5.0	Diesel	1.50	0.42	1665	1573	5290
Raised prereact temperature, C-free.		•	12	2.0	Diesel	1.50	0.43	1684	1676	2302
Comments (0¹)	8	Carbon Balance (Based on Exit Gas Comp.)	Reactor Pres- sure (PSIG)	Flow Rate (PPH)	Fuel	H ₂ O/C	0 ₂ /C	t Actual	mperatures (°F) Exit diabatic Cal- Flame culated	mperatur diabatic Flame
CATALYST LOADING: INLET 0.75 lbs PSD-3018 MID 0.53 lbs HGC-1030 EXIT 1.81 lbs PSD-2001	CATAL								•	,
INLET 0.75 lbs	CATA									

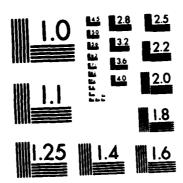
Carbon balance based on total gas flow exiting reactor measured with a wet test (W/T) meter.

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14B

Point Time Pre- (Hours) reaction





MICROCOPY RESOLUTION TEST CHART NATIONAL BUREAU OF STANDARDS-1963-A

INLET 0.75 lbs MID 0.53 lbs EXIT 1.81 lbs

CATALYST LOADING:

SUMMARY OF RUN 21 DATA POINTS

	Comments (0')	Increased temperature to look at fuel conversion. (0.962)	Lowered exit temperature, carbon-free. (0.938)	increased flows so W/T meter could be used to calculate the carbon balance at higher flows.		Readjusted fuel flow. (0.952)	Lowered temperature to look at fuel conversion. (0.938) Calibrate W/T meter, tried to correct previous results based on "as is" calibration.	Lowered flow, carbon formed. (0.934)	Raised temperature. (0.958)	First MeOH Point 3.48 PPH MeOH $\equiv 2.0$ PPH No. 2. (1.00) (Gas compositions look too low)	Lowered temperature gas composition still too low. (0.988)	Continued point, N_2 now looks high. (0.985)	Lowered temperature carbon balance low. (0.977)	Gas analysis finally correct. (0.984), (Carbon formed)
	8	0.881	998.0	0.910		0.927	0.912	988.0	0.936					
Carbon Balance (Based on	Exit Gas Comp.)	0.93+	0.92+	0.956+		0.956+	0.973+	0.947+	0.978+	0.972+	0.884+	0.972+	0.919+	1.02+
Reactor Pres-	sure (PSIG)	12	12	2	12	12	12	12	12	10.0	9.5	9.8	9.0	9.5
NO IL	_	1.5	1.5	2.	2.	5.0	2.0	1.5	1.5	3.48	3.48	3.48	3.48	3.48
Fuel	Ļ	Diesel	Diesel	Diesel	Diesel	Diesel	Diesel	Diesel	Diesel	Methanol	Methanol	Methanol	Methanol	1.90 Methanol
420/C		1.50	1.50	1.55	1.55	1.50	1.50	1.50	1.50	1.90 K	1.90 N	1.90	1.90 ¥	1.90 A
02/C H20/C		0.48	0.43	0.423	0.46	0.45	0.41	0.407	0.473	0.467	0.40	0.40	0.37	0.37
	Actual	1731	1666	1700	1766	1748	1675	1632	1729	1639	1515	1508	1434	1459
es (°F)	Cal- culated	1756	1635	1642	1762	1725	1609	1570	1728					
Temperatures	Adiabatic Flame	2450	2353	2356	2457	2439	2344	2303	2422					
1		1277	1271	1308	1328	1315	1299	1268	1257					
'	Fime Pre- (Hours) reaction	528	5 91	564	267	712	782	287	230	297	303	314	324	341
	Point Time (Hou	8	۲	23	23A	23B	54	52	8	23	28A	288	£	æ

+ Carbon balance based on total gas flow exiting reactor measured with a wet test (W/T) meter.

PSD-3018 HGC-1030 PSD-2001

INLET 0.75 Ibs MID 0.53 Ibs EXIT 1.81 Ibs

CATALYST LOADING:

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CONTROL AND CONTROL CONTROL CONTROL CONTROL CONTROL

SUMMARY OF RUN 21 DATA POINTS

·			1.90 (1.90 1.38 1.38 1.38 1.39 1.39 1.38 1.38 1.39 1.39 1.38 1.39 1.39 1.39 1.39 1.39 1.39 1.39 1.39	1.90 Methanol 1.90 Methanol 1.90 Methanol 1.90 Methanol 1.90 Methanol 1.78 Methanol Contam. with 58 Diesel	1 0 1	Reactor Pres- sure (PSIG) 9.5 10.5 10.5 16	Balance (Based on Exit Gas Comp.) 1.03+ 0.996 0.959	8	Raised exit temperature with increased air. (0.991) No gas analysis, AP stopped increasing. No gas analysis, raised O ₂ /C, AP still did not go down. Raised flows no H/C observed using FID of exit. (1.00) Flows too high to use W/T meter for carbon balance, but gas chromatograph has been reworked and N ₂ concentrations appear to be accurate. Lowered temperature with air, still C-free. (1.00) Temperature lower than desired, but preheat burner temperatures are very high. (0.978)
	1539 0	0.46	1.78	Methanol Contam. with 5° Diesel Methanol	5.22	91 71	0.930		Continued point, no decay. (0.977) Raised air (temperature). (0.984)
	1634 (0.46	1.78	with 5% Diesel Methanol Contam.	5.22	17	0.964		Continued point, no decay. (0.982)

+ Carbon balance based on total gas flow exiting reactor measured with a wet test (W/T) meter.

(3)

(E)

APPENDIX B

SUMMARY OF RUN 21 DATA POINTS

	Comments (8')	Lowered flow, did not increase conversion. (0.980)	Repeat of point 23B C-free. (0.956) NH ₃ analysis <10PPM.	NH ₃ <10 PPM, carbon formed.	C-boundary (0.959)		Carbon free at low $\mbox{H}_2\mbox{O/C}$. No gas analysis.	Carbon formed very slowly (0.927)	Carbon free (boundary) (0.934)	Carbon free at lower flow (0.934)	Carbon Free (0.938)	Carbon formed (0.933)	Carbon free H ₃ PO ₄ addition starting at 632 Hrs. (0.933)	Continuing point with acid addition, C-free (0.933)	Turned off acid right after this reading (0.931)	Carbon formed, no gas analysis	Carbon free	Carbon free, no gas analysis shut rig down.	
H	5		906.0		0.943			0.922	0.931	0.901	0.923	0.922	0.928	0.937	0.927			•	
Carbon Balance (Based on	Exit Gas Comp.)	0.986	0.95		0.983		,	0.995	0.996	0.965	0.984	0.991	966.0	1.004	0.997	•		•	
Reactor Pres-	sure (PSIG)	10.5	12.0	12.0	12.0		=	=	11.5	7.5	12.5	12.0	12.0	11.5	12.0	13.0	13.0	12.5	
<u> </u>	Rate (PPH)	3.48	2.0	5.0	5.0		2.0	5.0	2.0	1.5	2.0	2.0	2.0	2.0	2.0	5.0	2.0	2.0	
Fuel	_	Methanol Contam. with 58 Diesel	Diesel	Diesel	Diesel		Diesel	Diesel	Diesel	Diesel	Diesel	Diesel	Diesel	Diesel	Diesel	Diesel	Diesel	Diesel	
H ₂ 0/C		1.78	1.50	1.50	1.50		1.13	1.51	1.51	1.51	1.51	1.51	1.51	1.51	1.51	1.51	1.51	1.51	
02/C		0.44	0.45	0.40	4		9.4	0.45	4.0	0. 4	4.0	4.0	4.0	0.44	9.4	0.41	0.44	0.43	
	Actual	1545	1740	1660	1221	ection	1708	1671	1703	1692	1706	1686	1716	1703	1706	1648	1706	1688	
res (°F) Exit	culated		1682	1562	1670	Direct Fuel Injection	1682	1630	1688	1704	1695	1675	1715	1715	1715	1630	1693	1665	
Temperatures (°F	Adiabatic Flame		2389	2300	2385	Dir	2544	2430	2474	2496	2484	2465	5052	2508	5208	2440	2482	2460	
	reaction		1268	1278	1282		1227	1242	1242	1263	1252	1234	1275	1274	1275	1271	1250	1249	
	<u>ي</u>	3	475	518	929		3	8 28	286	294	619	632	675	710	752	753	174	782	
	Point Time (Hou	%	93	\$	Ŧ		2	\$	\$	\$	\$	41	8	6	33	51	25	23	

APPENDIX C

EVALUATION OF A MIXED GAS CYCLE FUEL CELL SYSTEM USING AN ADIABATIC REACTOR

COMPATIBLE WITH MILITARY LOGISTIC FUELS

TASK 3

SYSTEM ANALYSIS AND COMPONENT DEFINITION

Leslie Van Dine

March 22,1983

TASK OBJECTIVES

Define a system configuration for a mixed gas cycle (MGC) using Diesel fuel

a 1.5 KW mixed gas cycle system: Estimate the characteristics of

* Efficiency
* Weight
* Volume

Extrapolate characteristics to larger powerplant ratings

* Up to 100 KW

Identify key technology areas and recommend potential system improvements

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MAJOR STUDY CONCLUSIONS

- oil. * A mixed-gas-cycle fuel cell system is thermodynamically practical and capability including high sulfur diesel Multifuel offers the benefits of:
 - A 1.5 KW mixed-gas cycle fuel cell powerplant is heavier, larger, and less efficient than the 1.5 KW Army methanol fuel cell system. No system water requirement.
- * Higher power ratings dramatically reduce sysytem weight and volume

: 0 0	FT3/KW	-40%	-55%	-59%
EFFECT	LBS/KW	-45%	-54%	-56%
CHANGE IN	RATING	1.5> 10 KW	1.5> 40 KW	1.5> 100 KW

- * Desulfurization requirements add from 26 to 45% to system weight depending on rating.
- * Special areas of technology concern unique to the MGC include: High temperature heat exchangers
 - Anode recycle gas pump

 - Acid management
 - Ammonia removal

1.5 KILOWATT MIXED GAS CYCLE F/C SYSTEM

ACCUS CONSIGNACE CONTRACTOR PROSESSION PROGRESSION AND ACCUSED TO

Residence of the second of the

GROUNDRULES

Fuel processor operating conditions and characteristics based on the experimental program.

Minimum change from the Army 1.5 KW Methanol fuel cell design.

System weights, volumes, and efficiencies calculated for powerplant ratings from 1.5 to 100 kilowatts.

Fart power, transient response, and start-up outside the scope of this study.

1.5 KILOWATT MIXED GAS CYCLE FUEL CELL SYSTEM KEY ASSUMPTIONS

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Fuel:

Diesel oil - CH1.8 Higher heating value - 19500 btu/lb 5000 ppmw sulfur

Fuel Cell: Based on the Army 1.5 Methanol Fuel Cell

Cooled by recycling anode fuel gas 1 ATM operating pressure 340 DEGF average cell temperature .283 ft2 active area / cell 90 watts/ft2 baseline cell performance 90% H2 utilization (internal 23%) 70% O2 utilization

1.5 KILOWATT MIXED GAS CYCLE FUEL CELL SYSTEM KEY ASSUMPTIONS (CONTINUED)

Fuel processing: Based on experimental tests of the adiabatic reactor for the mixed-gas-cycle

1600 DEGF exhaust temperature .4 moles O2 per atom of fuel carbon 1.82 moles H2O per atom of fuel carbon Space velocity = 12 # fuel/(hr-ft3 catalyst)

Desulfurization:Disposable ZnO beds

Sulfur slip = .01 ppm (Based on wet gas volume)

Ammonia scrubber:H3PO4 acid-charcoal bed

NH3 leaving reactor < 10 ppmv NH3 to fuel cell < .5 ppmv

Shift Converter:Low temperature catalyst

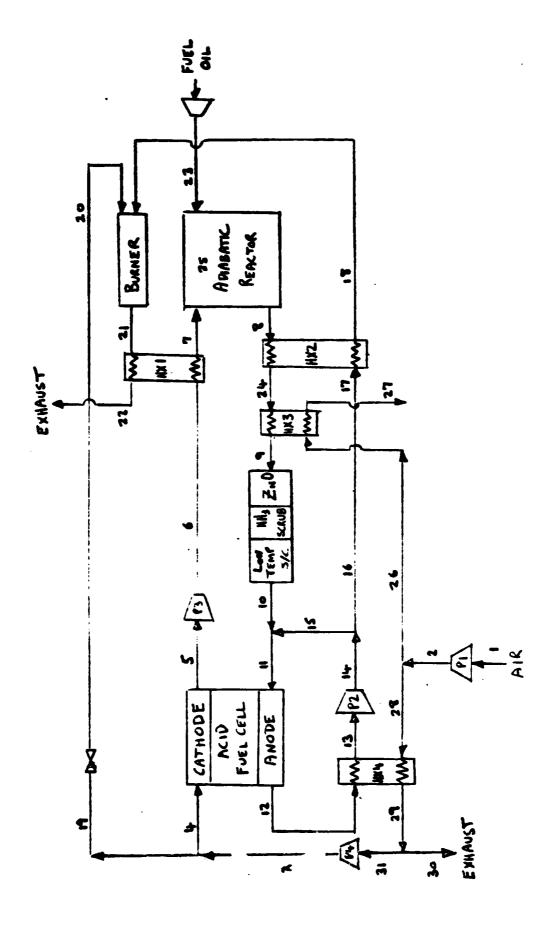
 $CO \ slip = 1 \% \ (Dry \ gas)$

Inverter efficiency = 85% (1.5 KW size)

Pumps-blower efficiencies scaled from commercial hardware specs.

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6



SYSTEM THERMODYNAMIC TABLE I.S KW

BTU/HR	HA44 HW444 HW644	11.000.	-1132 -2203 -2203	-73752 -73752 -75469	-753419 -728859	-2536 -2536 -1698		1-1-1-1-1-1-1-1-1-1-1-1-1-1-1-1-1-1-1-	-1978	7474 7474 7470	いい ひ44 いいぬ いいぬ
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EFFICIENCY COMPARISON 1.5 KW RATING

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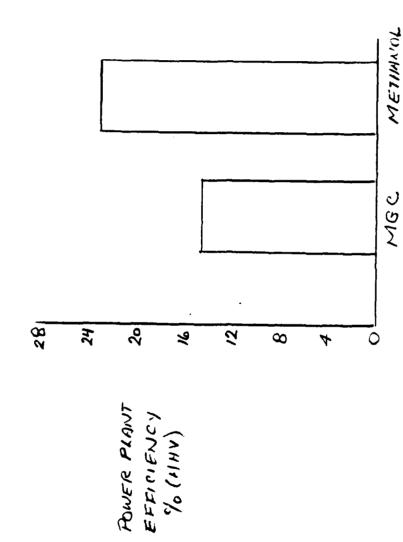
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33%

- * Relatively low efficiency of MGC system caused by:
- Parasite power associated with gas cooling
- Low pump, fan and motor efficiencies in small size
- Pressure drop associated with added components



MAJOR COMPONENTS ADDITIONS REQUIRED FOR DIESEL FUEL USE

■ こうかんしょう | 「日本シャンクション・「日本のののなって」 | 「日本ののできない。

1.5 KW POWERPLANT

- * Adiabatic reactor
- * Two high temperature heat exchangers
- * Shift converter
- * Ind desulfurization bed
- * Ammonia scrubber bed
- * Fuel cell coolant to air heat exchanger
- * Coolant pump (Gas containing hydrogen)
- * Larger support structure

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SPECIFIC WEIGHT COMPARISON

177

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12.22 May

F/P BASE 9 STRUCT CELLS B.O.P. . ≥ . BASE & STRUCT B. O. P. 2 NO CELLS 2/5 . .≻.<. FIP 6001 500 400 300 200 00/ SPECIFIC WEIGHT 185/KM

70MHL3H

MGC

C-11

VOLUME COMPARISON 1.5 KW PPLT. SPECIFIC

AND THE PROPERTY OF THE PROPER

NOTE

FOR MGC - PACKAGING FACTOR

OF 2 MAS USED ON

MAJOR COMPONENTS

20

ZNO STRUCT 77 4 9 80/ 9/ CPACKNSED) SPECIFIC VOLUME fr3/xw

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METHANCE

HGC

当时, 1966年, 1967年, 1967年, 1968年, 1968

IMPACT OF LARGER POWERPLANT RATINGS

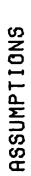
GROUNDRULES

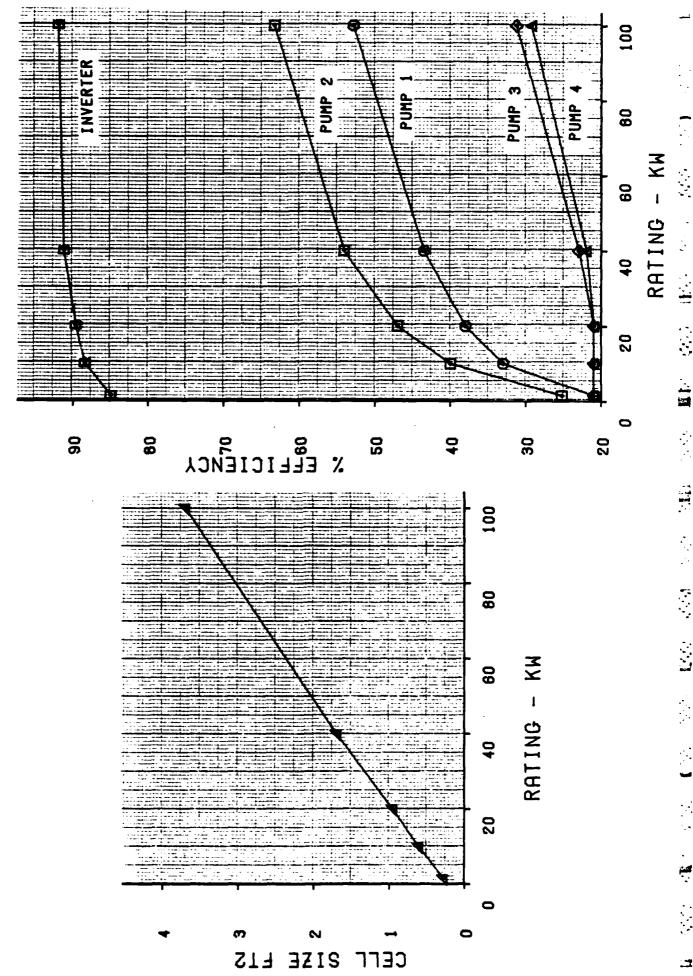
No start-up equipment included . (Assumes external start with electric heaters) Larger pumps, blowers, and motors with improved efficiency characteristics. Shift converter, and 2nO beds sized by computer modeling programs.

a space velocity derived from Adiabatic reactor sized by experimental tests.

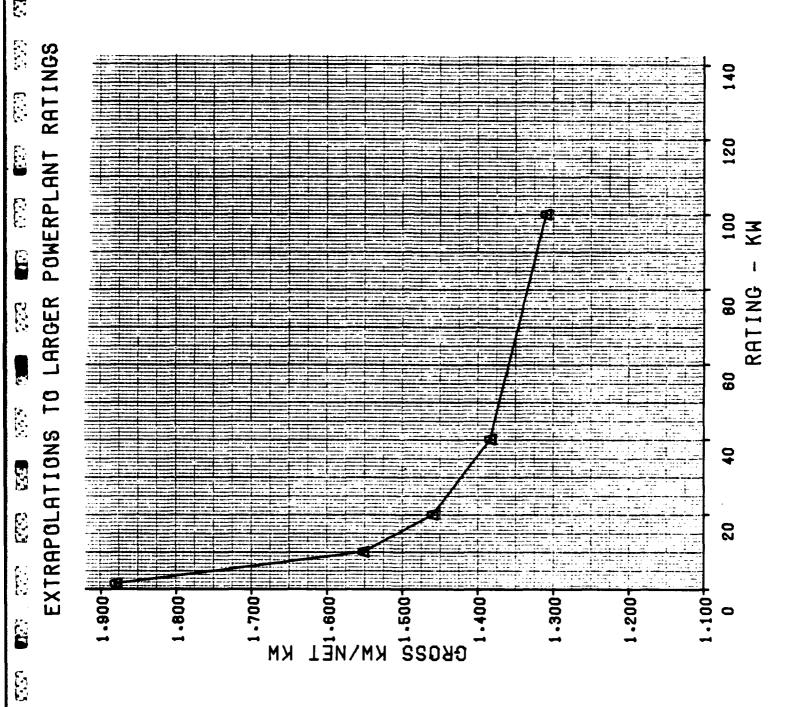
was used to account for controls,piping,valves,base and structure. Scaling was performed on other major components while a factor

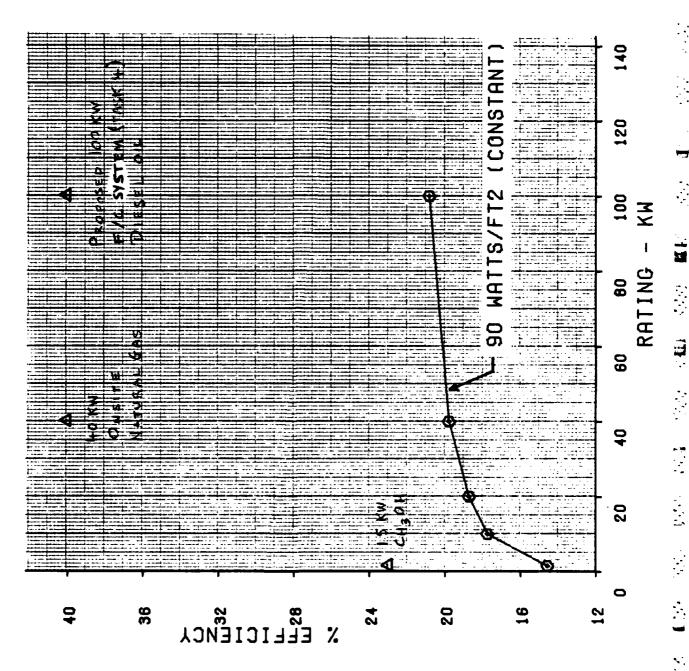
Cell size was increased at higher ratings. (Size based on previous utility studies)





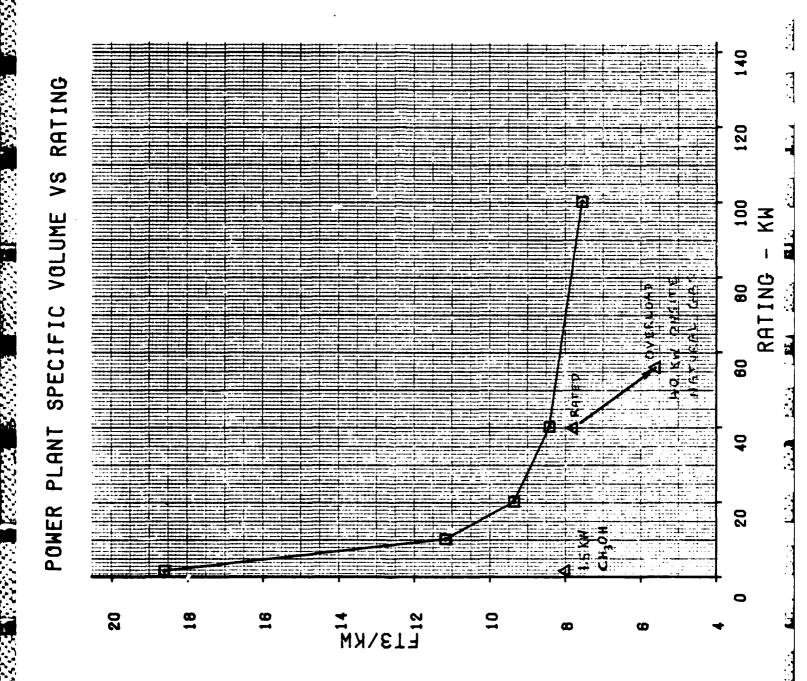
C-14





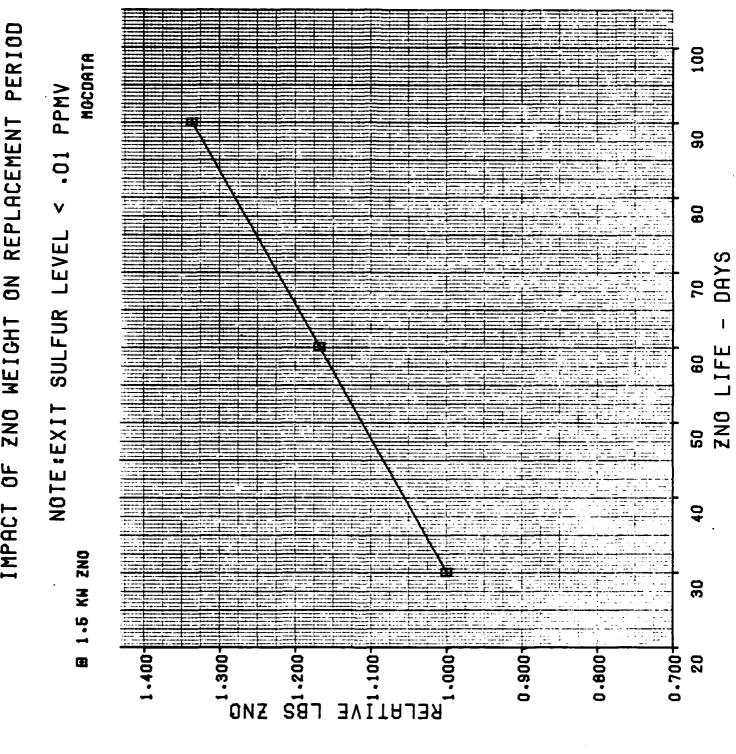
POWER PLANT SPECIFIC WEIGHT VS RATING 120 7 ± 00 X RATING 4 20 ES/KM 800 700 600 300 200 100

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IMPACT OF VARIATION IN POWER DENSITY 1.5 KW POWER PLANT 70 90 WATTS/FT2

IMPACT OF ZNO WEIGHT ON REPLACEMENT PERIOD



ALTERNATE SYSTEM OPTIONS

DIESEL FUEL - MIXED GAS CYCLE

- * Separate air cooling
- Reduced parasite power
- Elimination of cel) waste heat exchanger
- * Liquid two-phase dielectric cooling - Minimum parasite power
- CYCLIC REFORMER (BASELINE TASK 4 SYSTEM) DIESEL FUEL
- * Provides maximum efficiency
- * Requires water recovery for fuel processor operation

CRITICAL SYSTEM COMPONENTS

CANADASA (CORENTALISMOS COMPANIOS CONTRALISMOS CONTRALISM

* High temperature heat exchangers :

Max Gas Temp - DEGF	1800	1600
Effectiveness	78 %	% 68
	Preheater	Cooler
	Air	Gas
	Reactor	Process

- * Coolant recycle pump H2 bearing gas at 280 DEGF
- * Carbon free fuel vaporization with heated cathode exhaust
- * Impact of acid vapor corrosion
- * Size of ZnO beds
- * Effectiveness of ammonia scrubber

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RECOMMENDATIONS FOR FURTHER STUDIES

MIXED GAS CYCLE

- * Evaluate alternate system options
- * Select best option and perform following tasks
- Conceptual design of larger system
- Definition of performance and physical characteristics More detailed critical component definition
 - Consideration of:

Part power

Start-up requirements and time

Transient operation